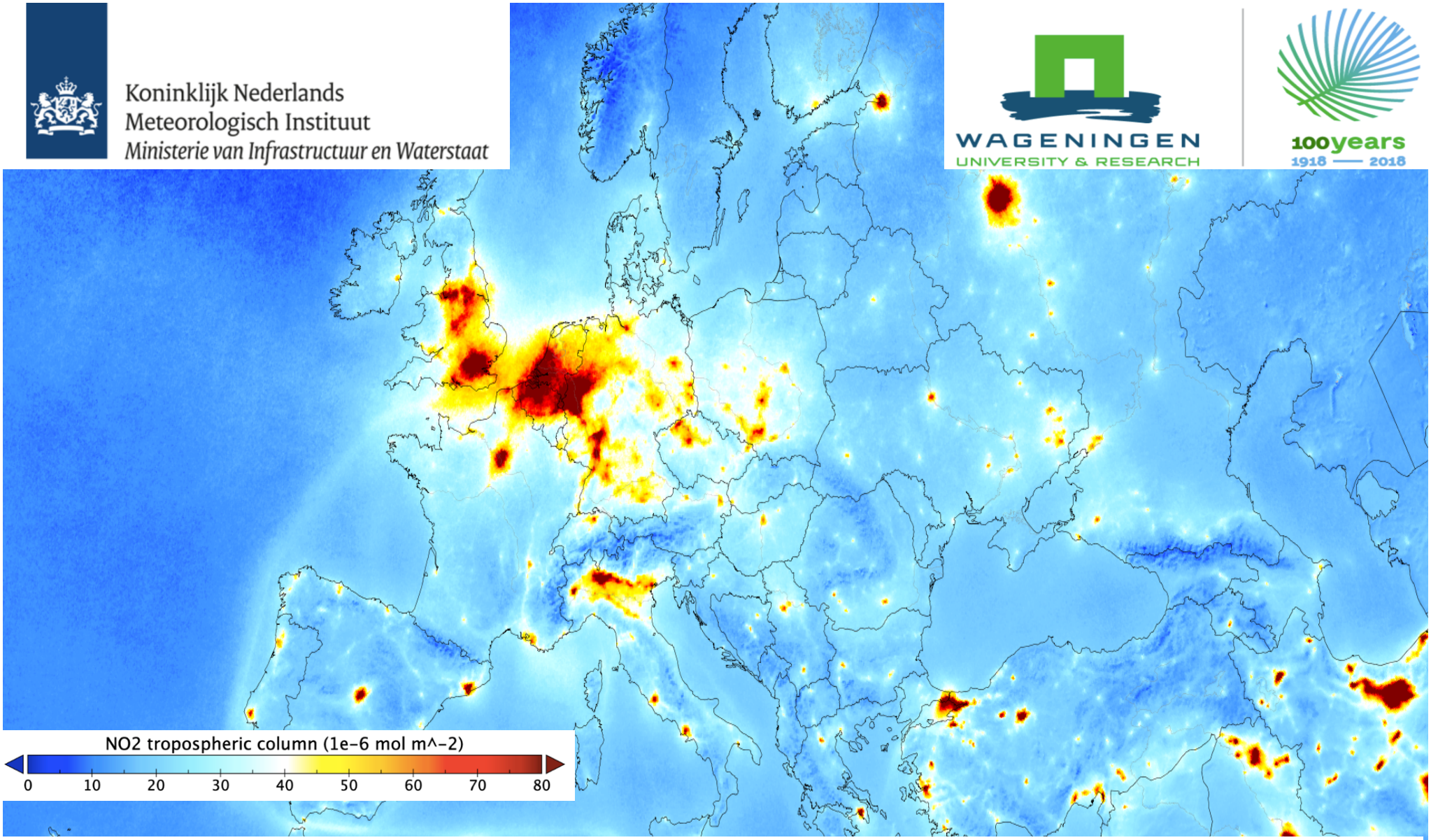
Koninklijk Nederlands
Meteorologisch Instituut
Ministerie van Infrastructuur en Waterstaat

WAGENINGEN
UNIVERSITY & RESEARCH

100years
1918 — 2018

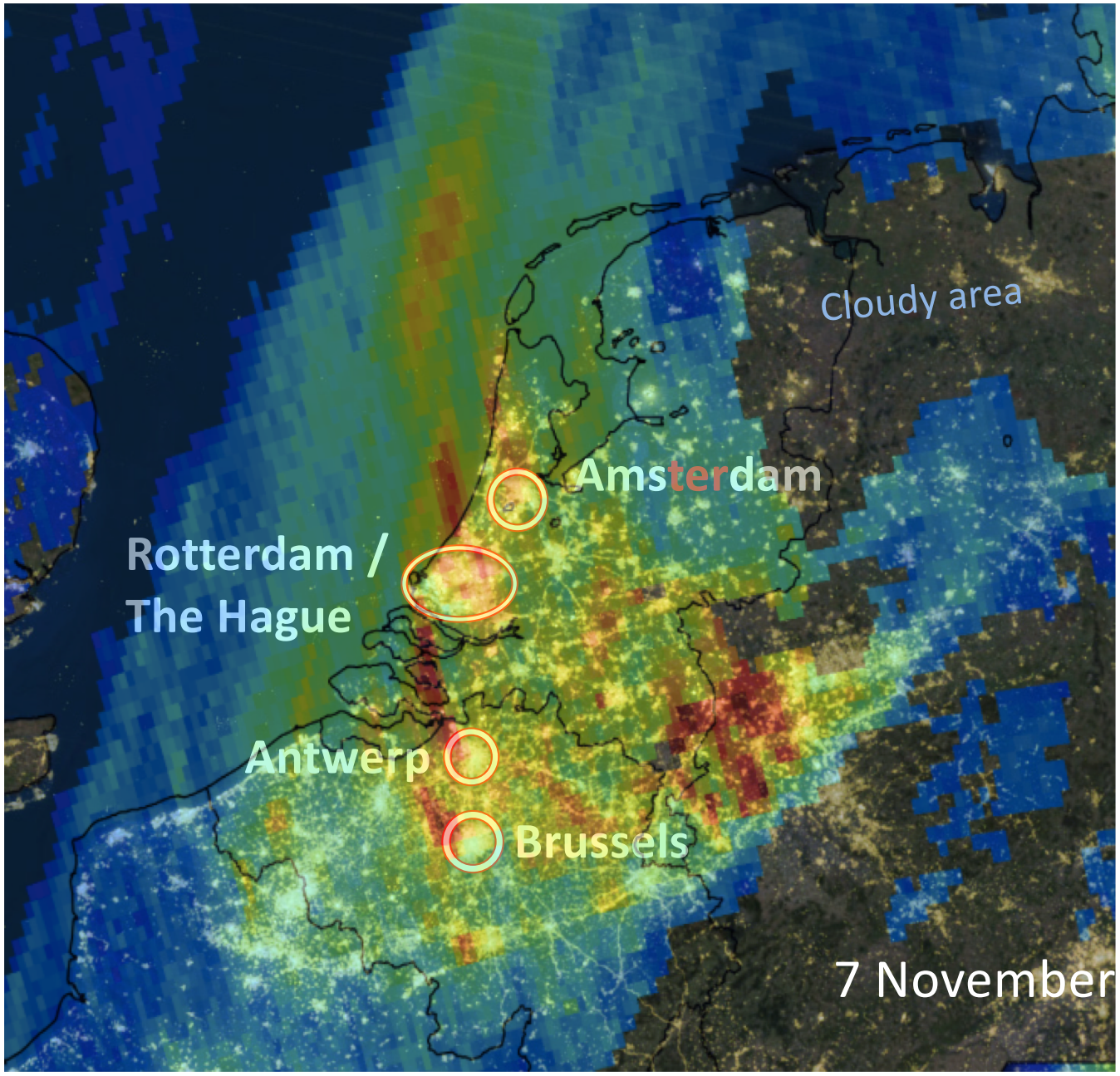


New perspectives on changes in tropospheric NO_2 from the OMI and TROPOMI sensors

Folkert Boersma

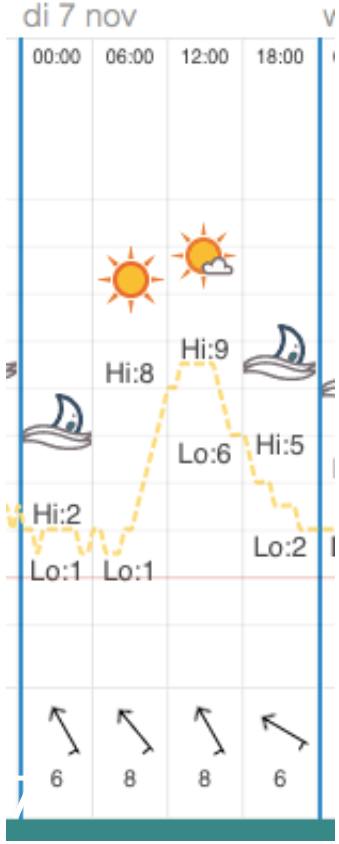
Alba Lorente, Henk Eskes, Pepijn Veefkind, Jos van Geffen, Maarten de Zeeuw, Hugo Denier van der Gon, Steffen Beirle, and Maarten Krol

NO_2 plumes in the Benelux



Right after instrument cooler opened

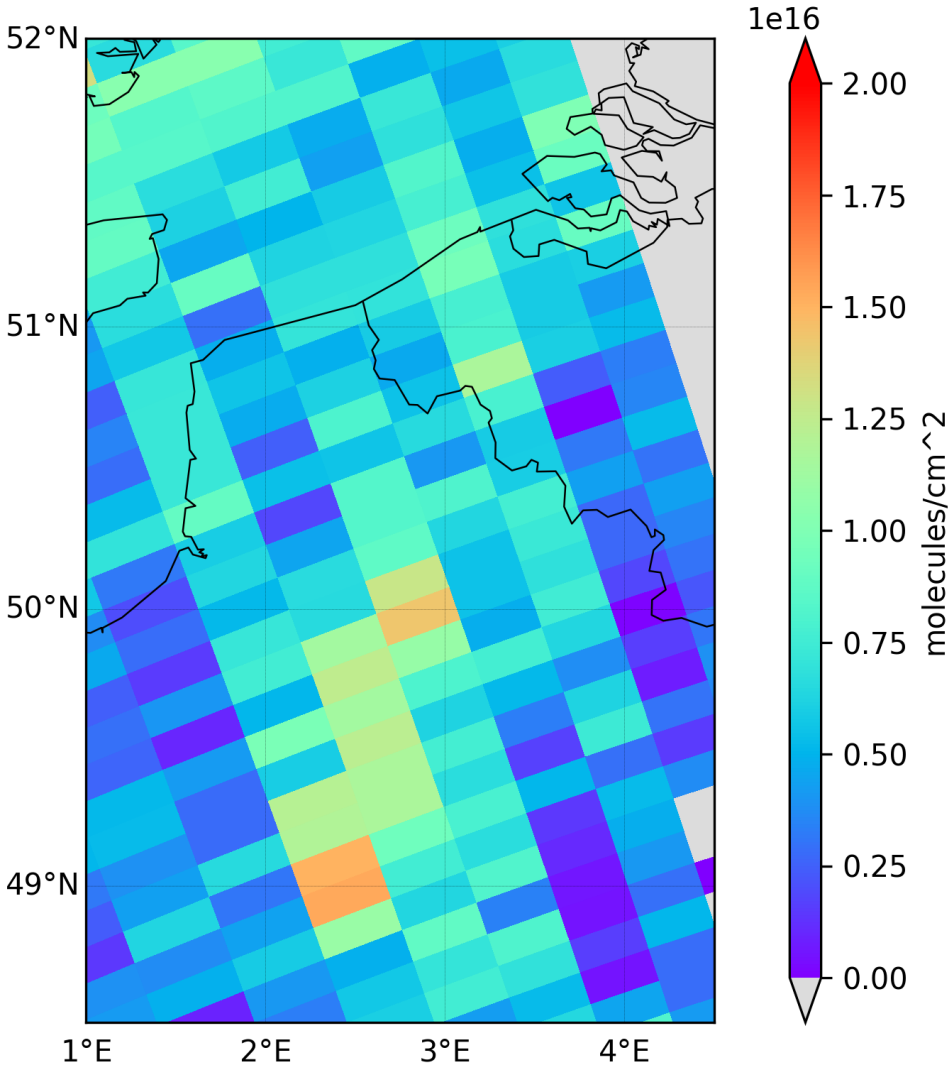
No measured solar irradiance spectrum yet



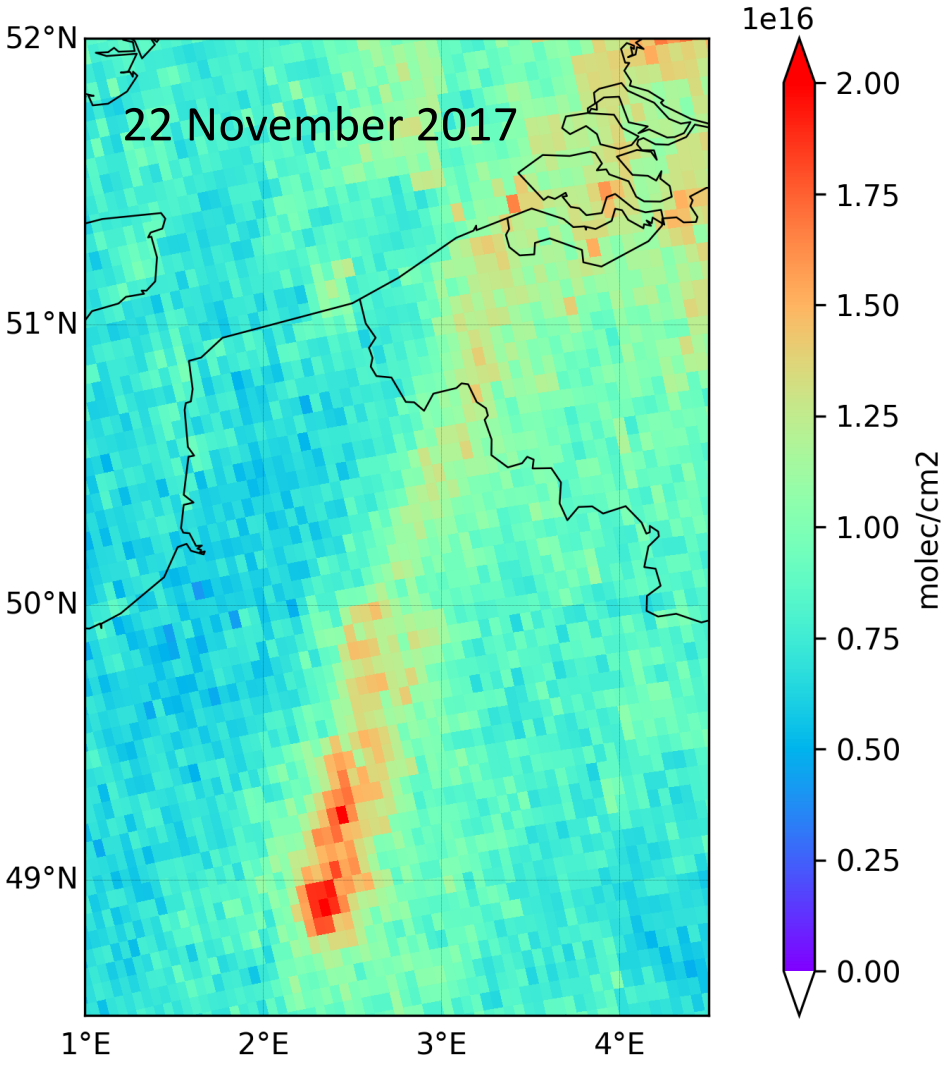
VIIRS nighttime lights

NO_2 plumes from Paris

OMI

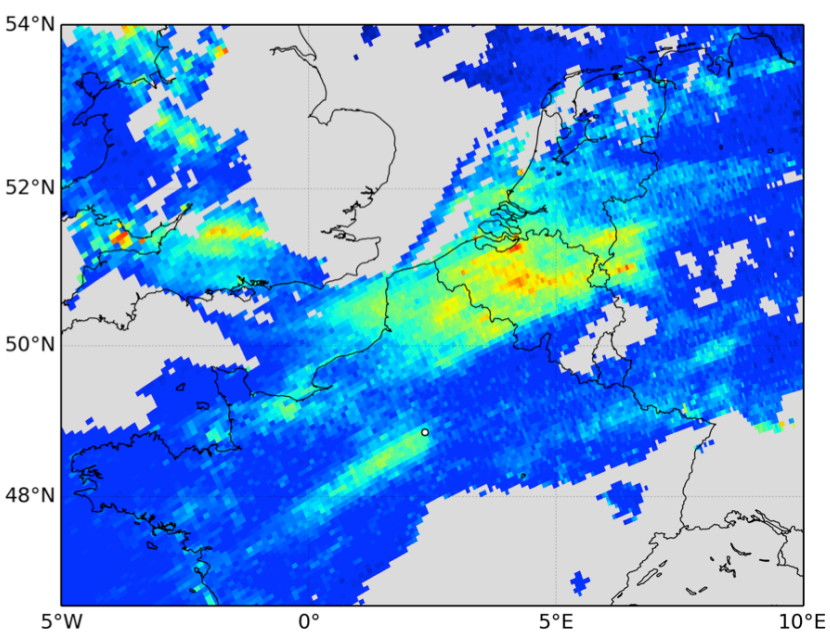


TROPOMI



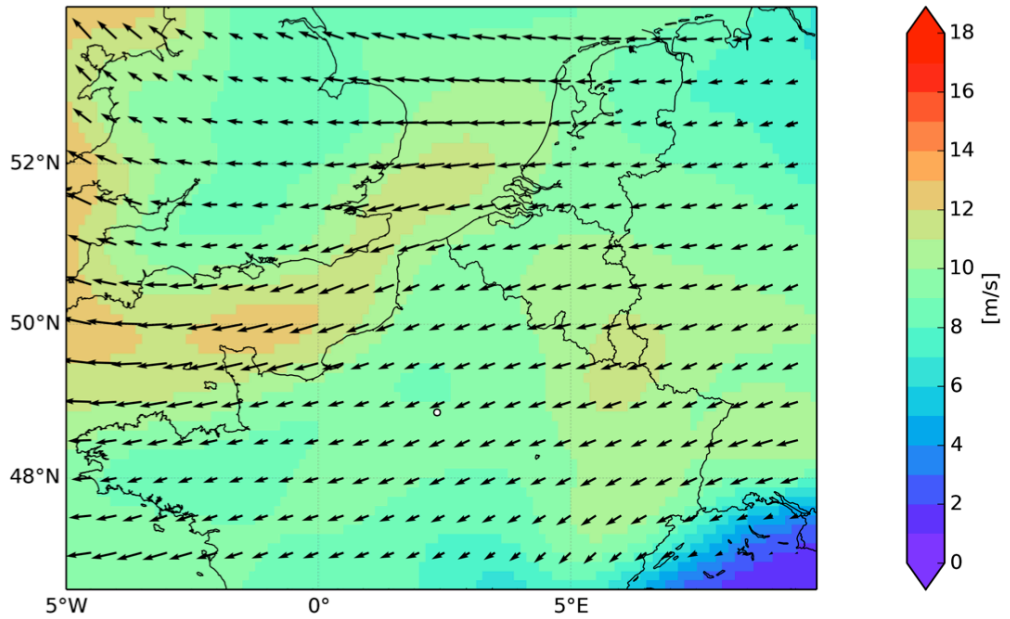
NO_2 plumes from Paris

TROPOMI NO_2 tropospheric VCD



23 February 2018

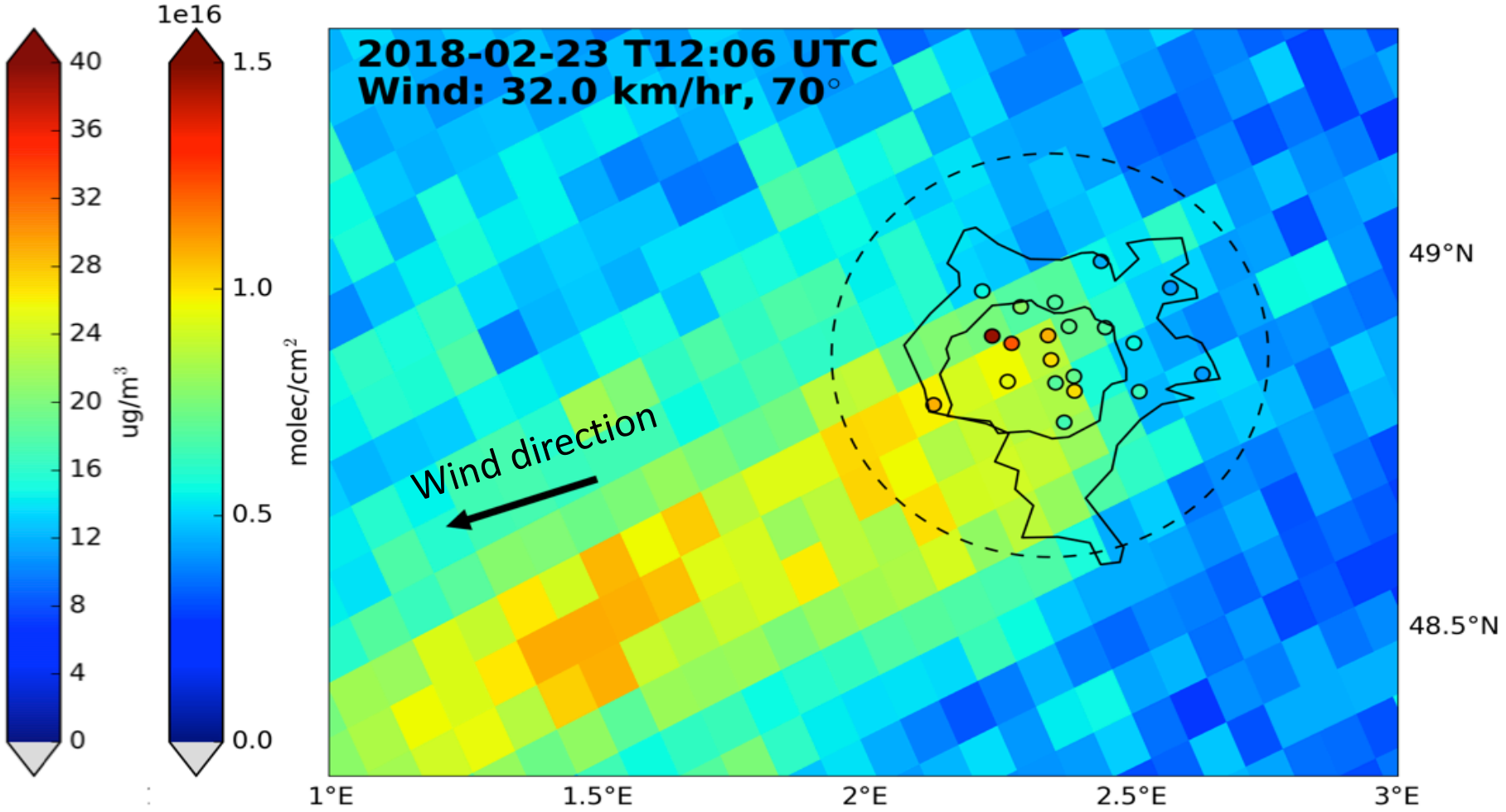
ECMWF ERA-I wind speed @BL



Wind at 12 UTC

NO_2 increase over Paris

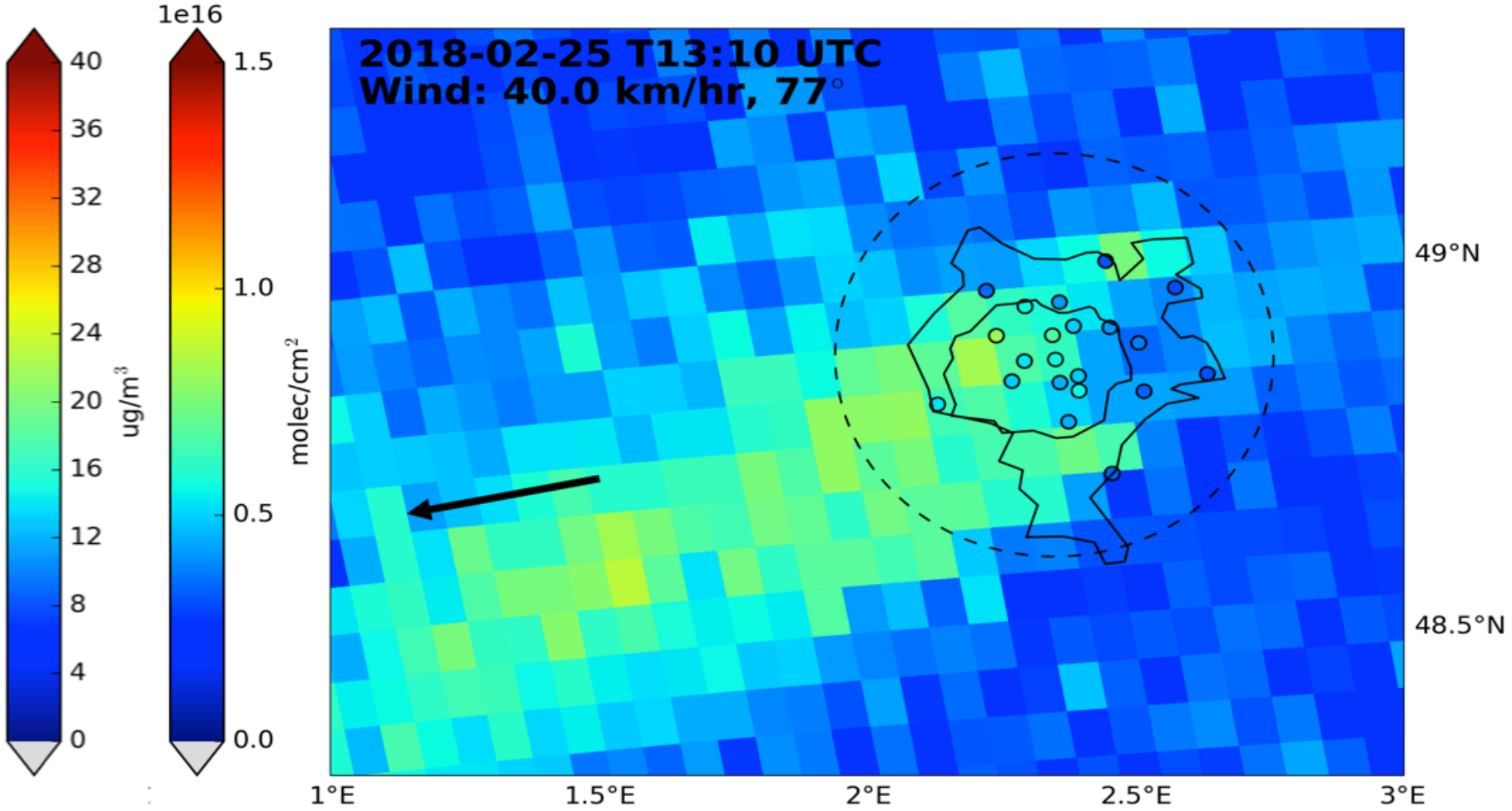
‘urban background’ surface measurement



FRIDAY

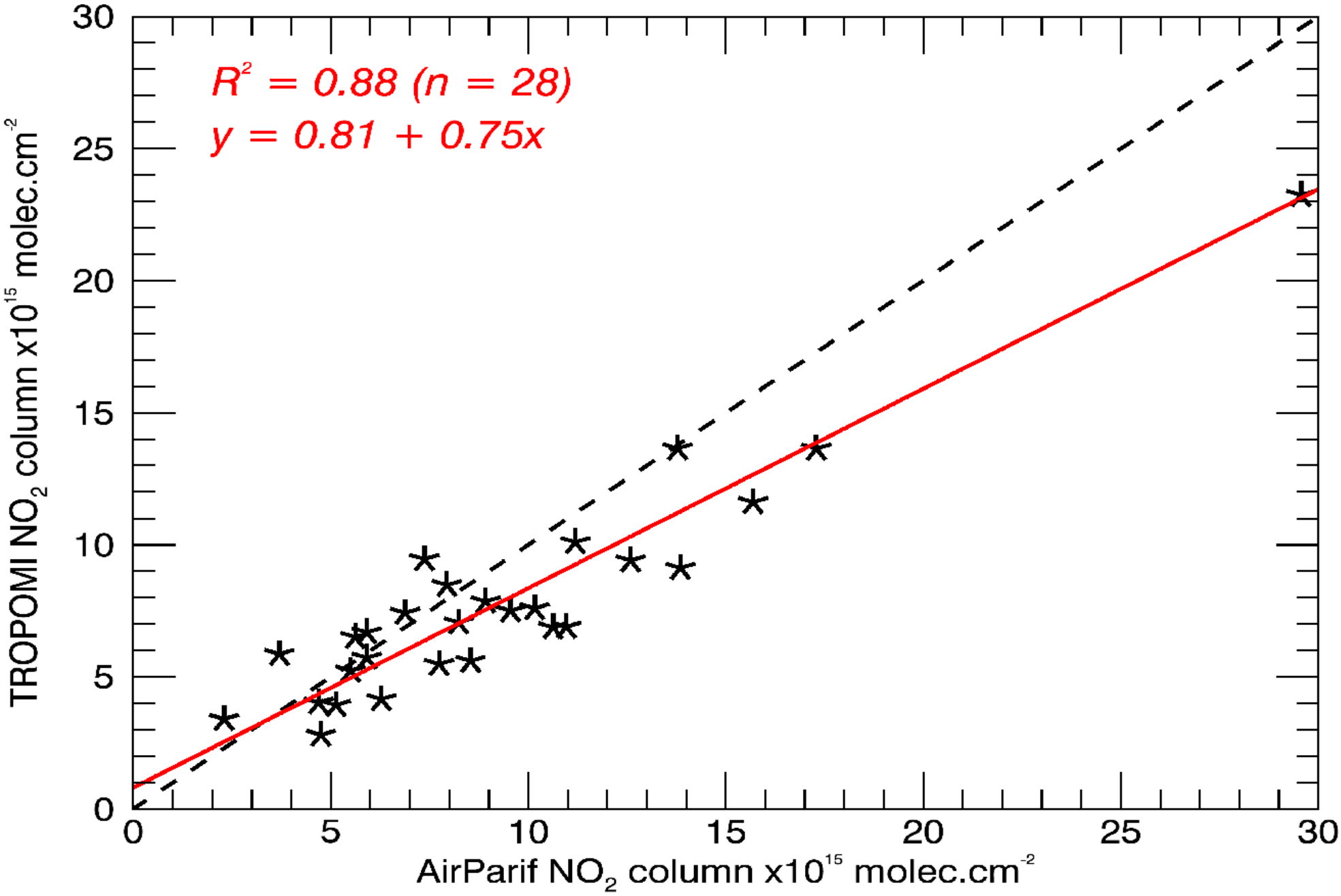
NO_2 increase over Paris

‘urban background’ surface measurement

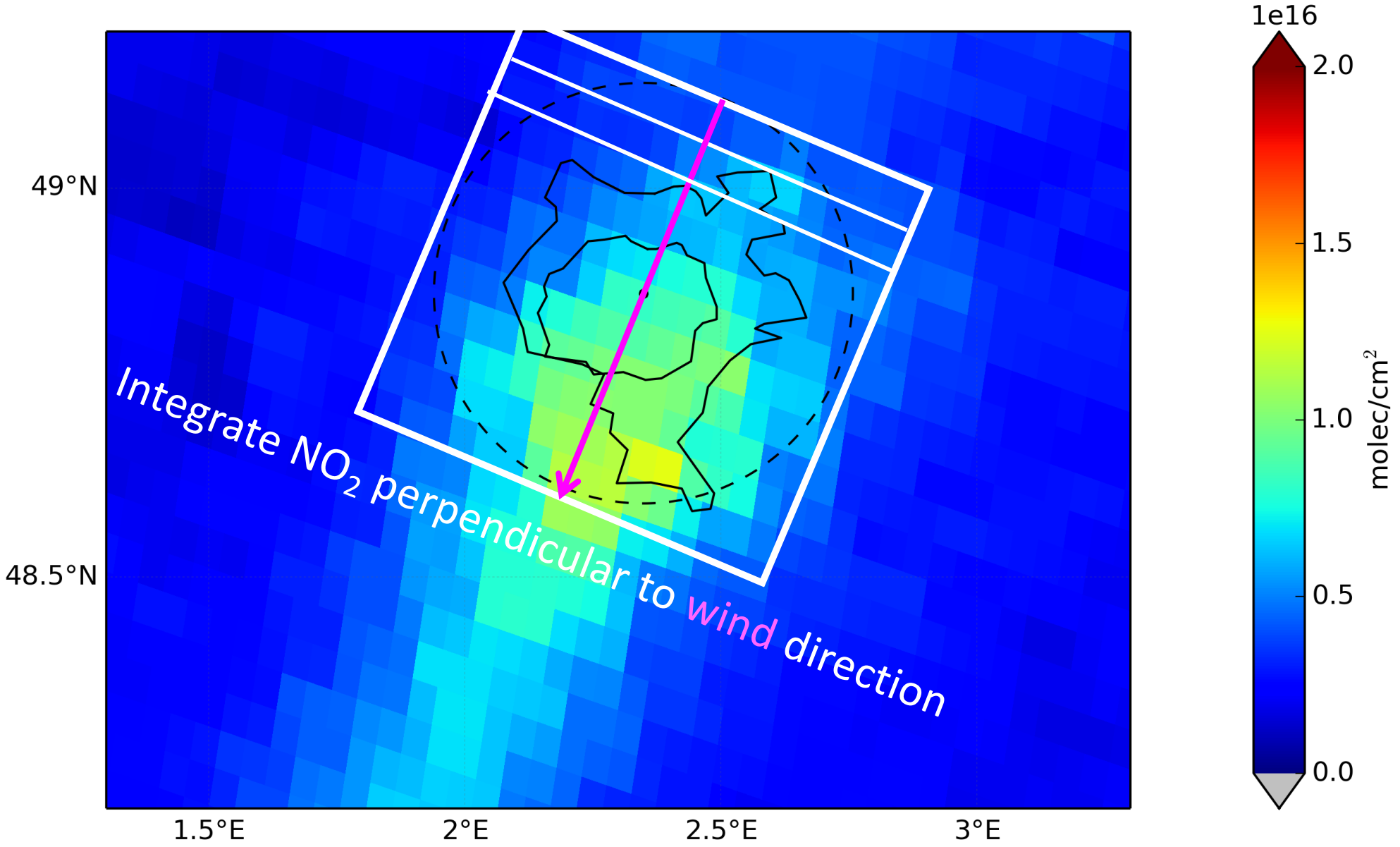


SUNDAY

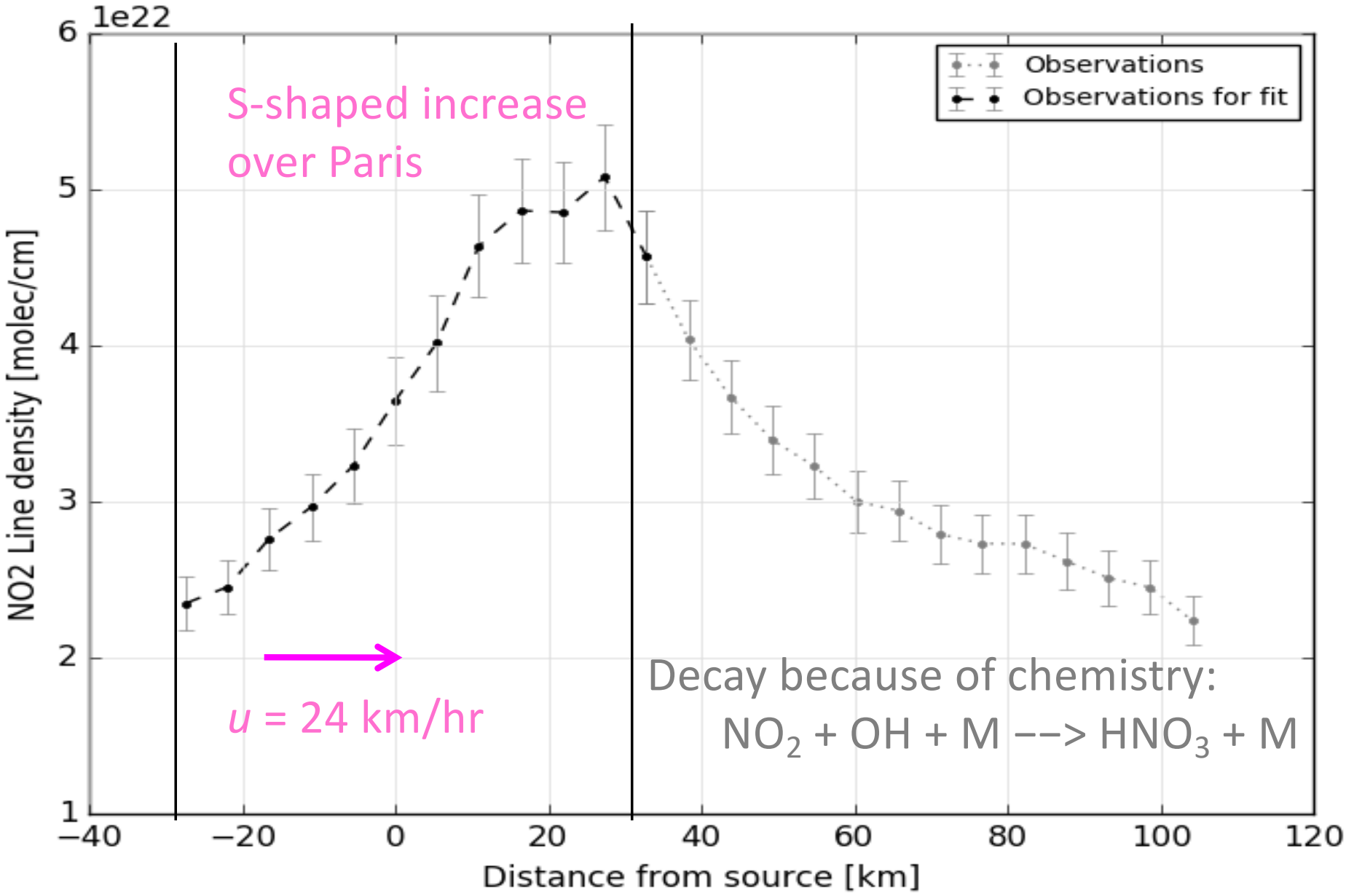
Validation of TROPOMI v1.1 at Eiffeltower



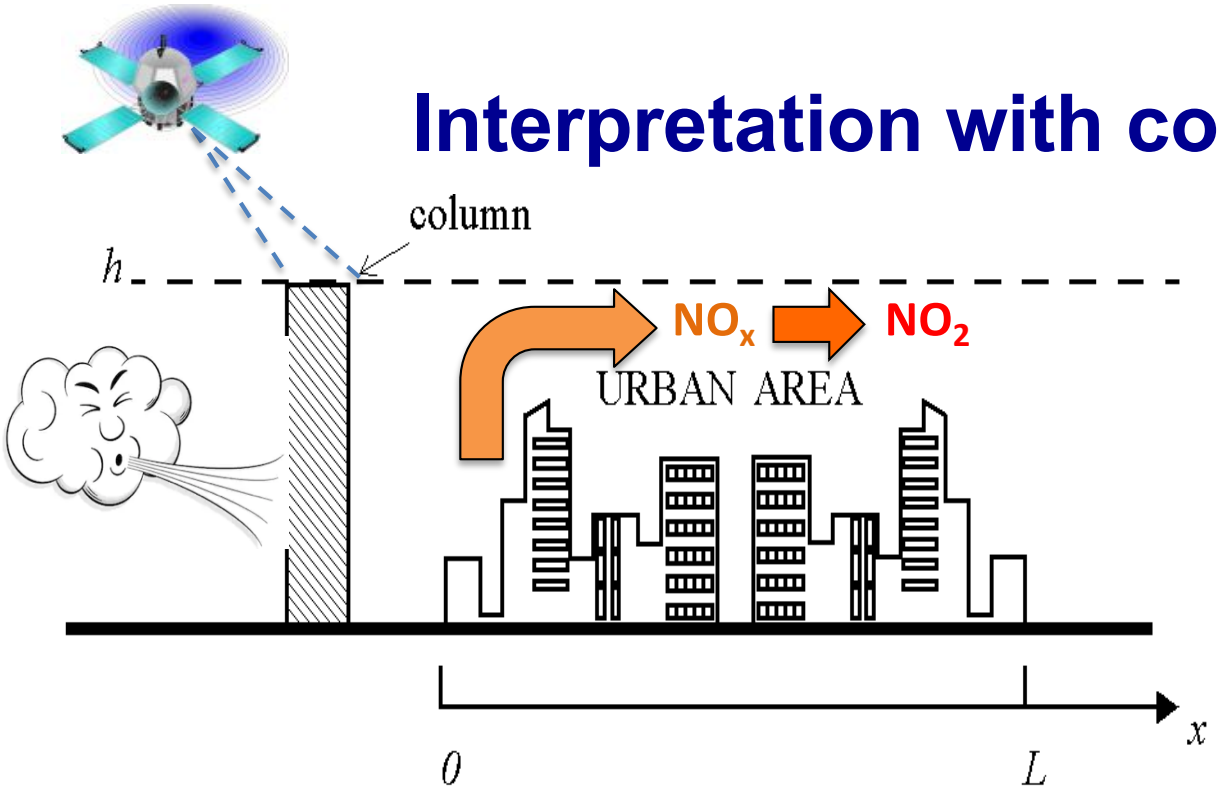
NO_2 line density over Paris



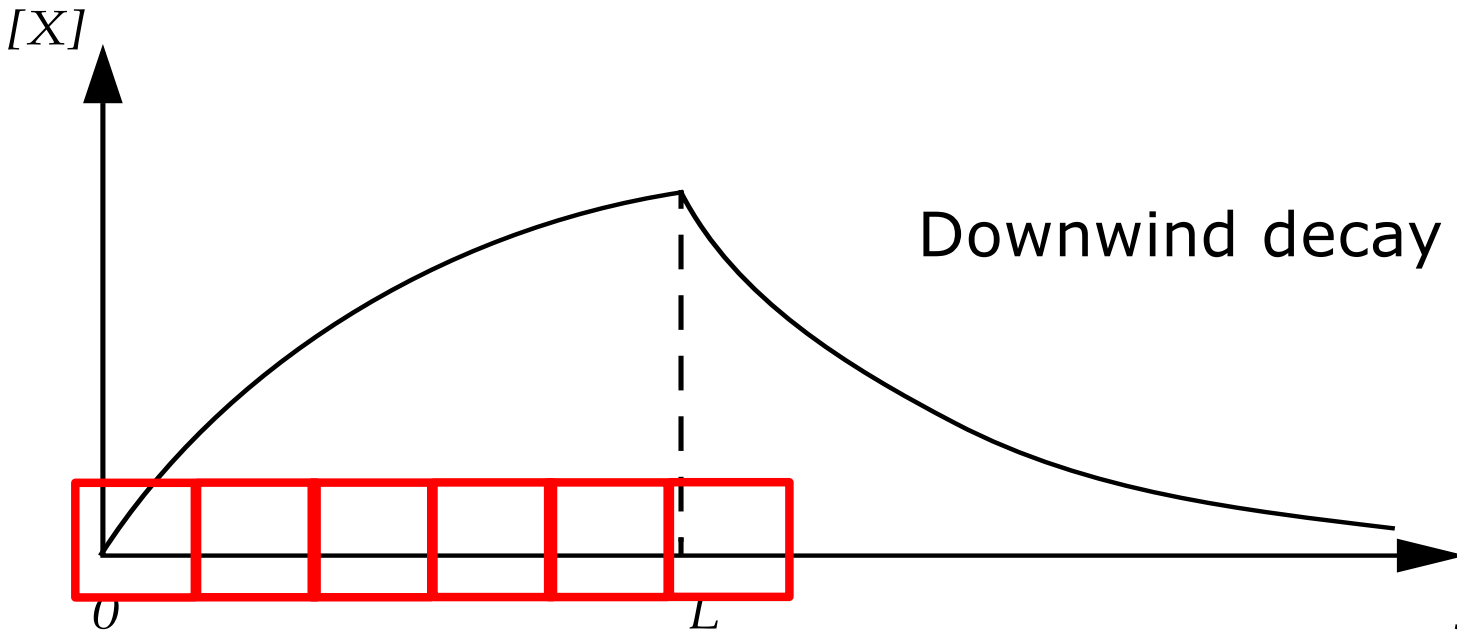
NO_2 line density over Paris



Interpretation with column model



$[NO_2]$ increases over the city



Downwind decay

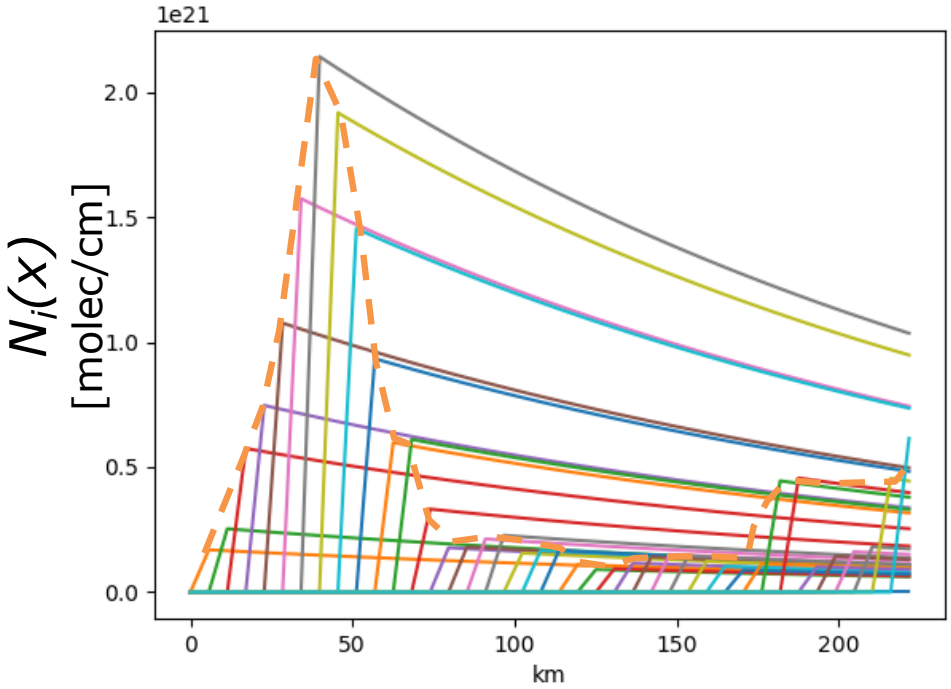
Emission pattern matters

Simulate line density by accounting for spatially varying emissions

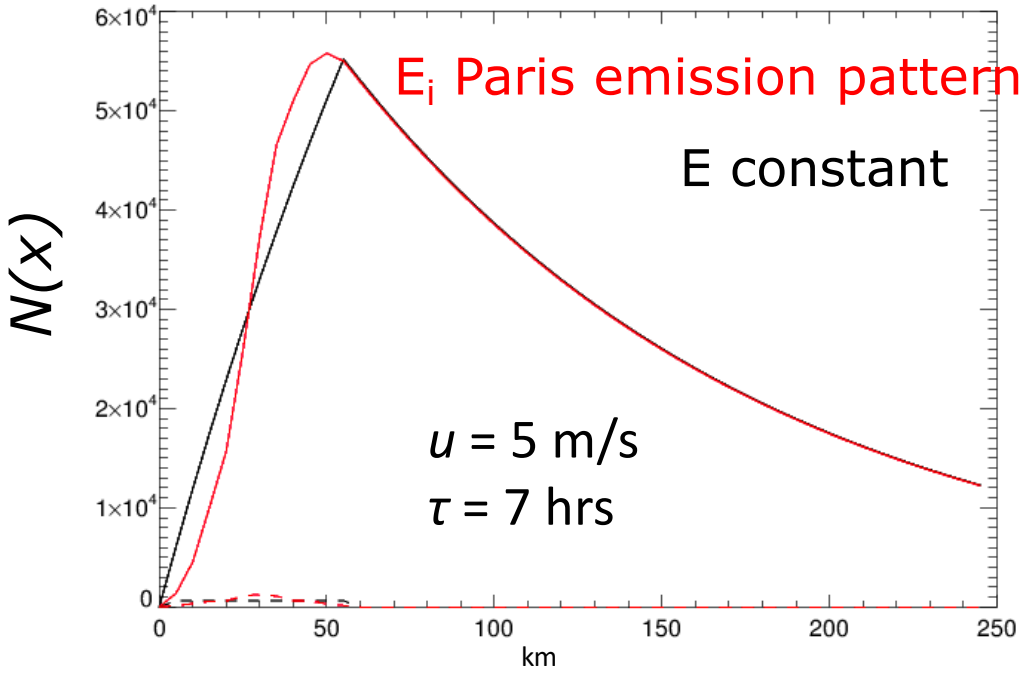
$$N_i(x) = \frac{E(x_i)}{k}\left(1 - e^{-k(x-x_i)/u}\right)\frac{[NO_2]}{[NO_x]} \quad \text{for } x \geq x_i$$

$$N_i(x) = 0 \quad \text{for } x < x_i$$

$$N(x) = \sum_{i=1}^{n} N_i(x)$$



TNO emission pattern - - - - -



Superposition model

Large ensemble of modeled line densities

Fixed parameters

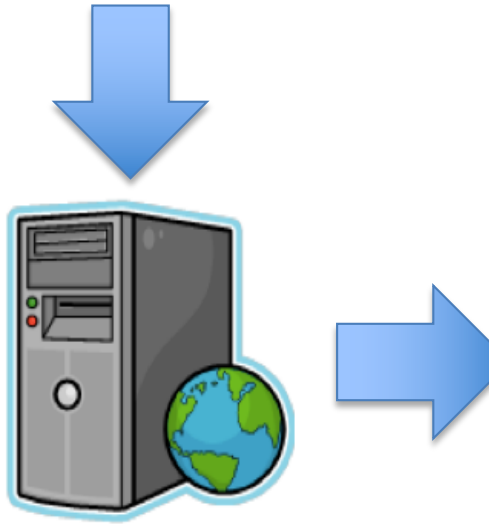
u boundary layer mean ECMWF wind

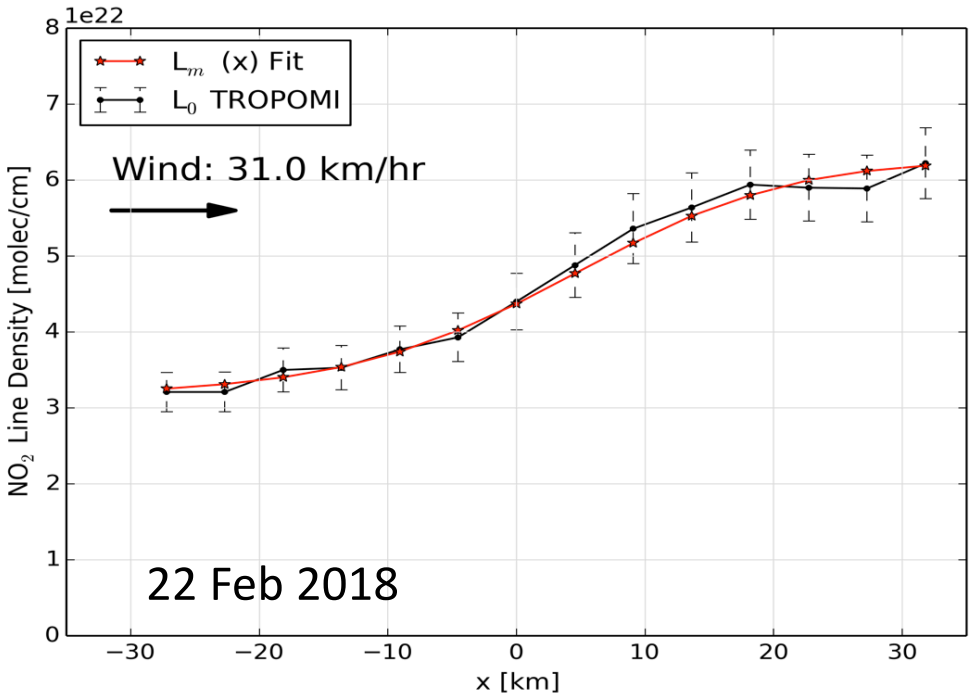
NO_x:NO_2 boundary layer mean from CAMS model

Varied parameters (5+1)

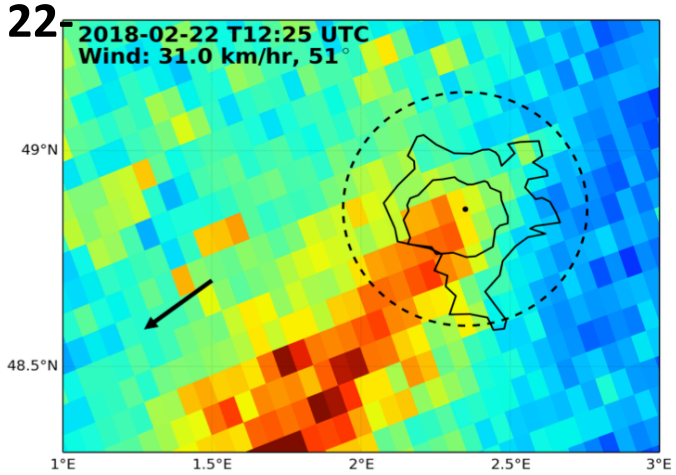
E_i – initial guess from TNO-MACC-III inventory

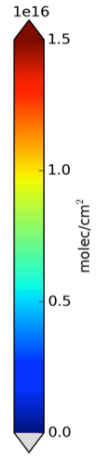
k – initial guess from CAMS model, uncertain because of [OH]

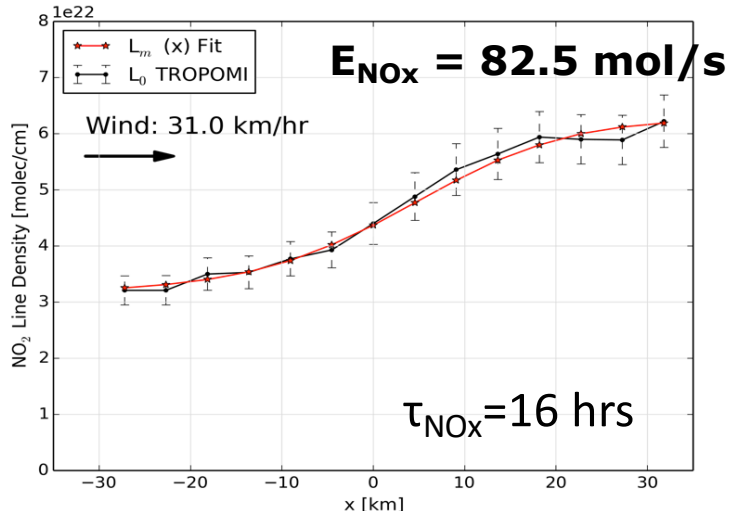




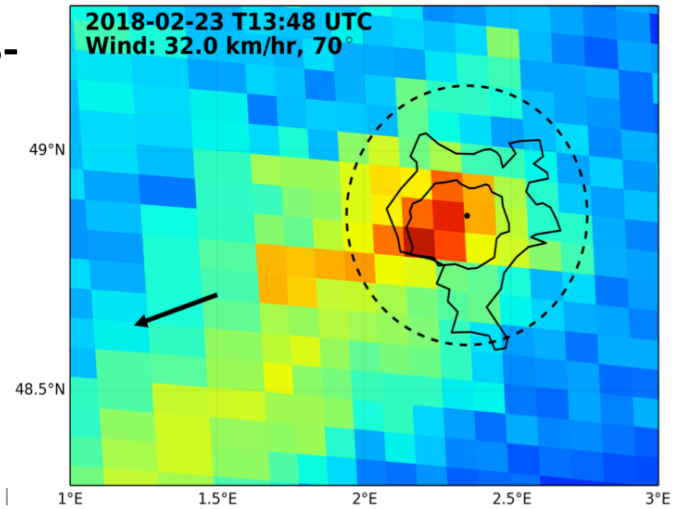
Thursday 22-02-2018
31 km/h

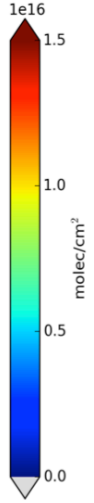


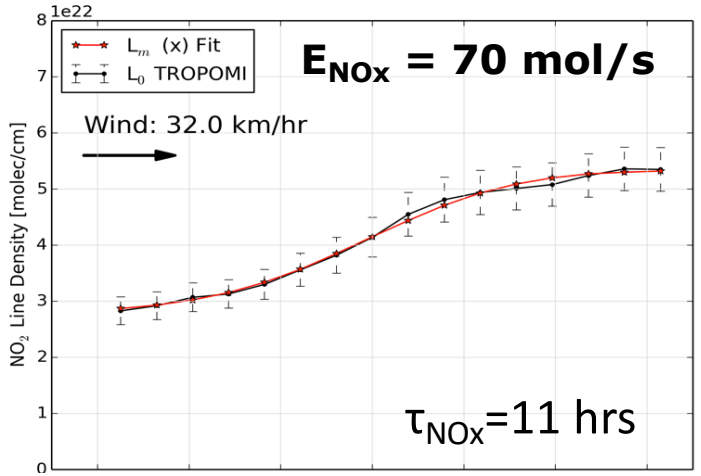




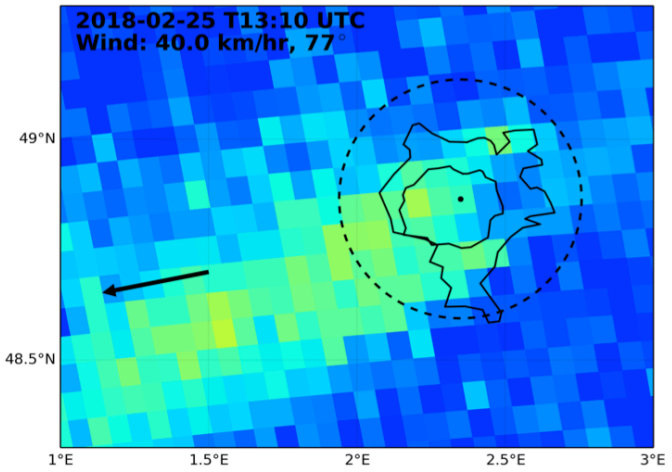
Friday 23-02-2018
32 km/h

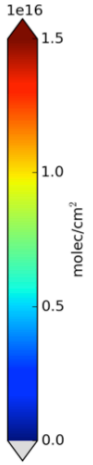


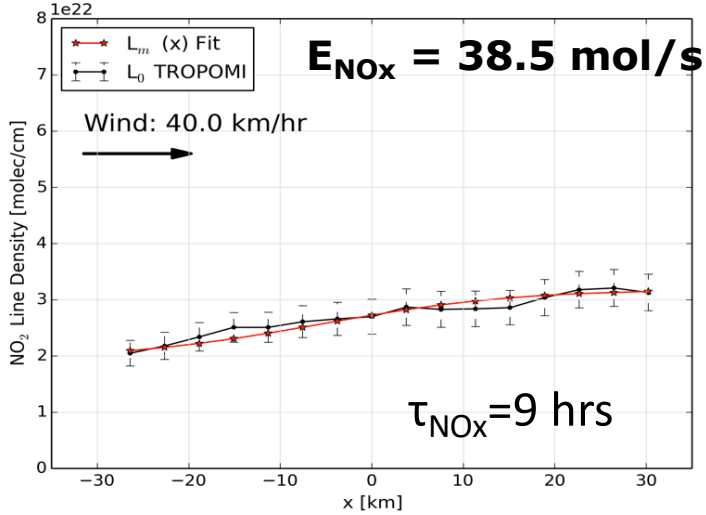




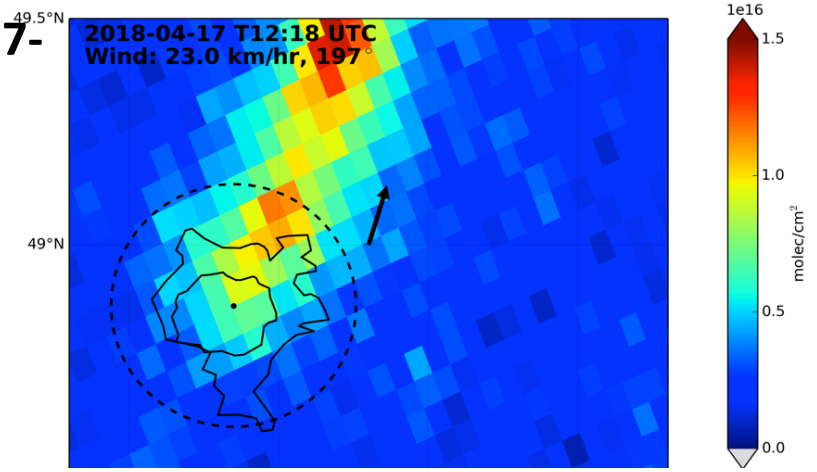
Sunday 25-02-2018
40 km/h

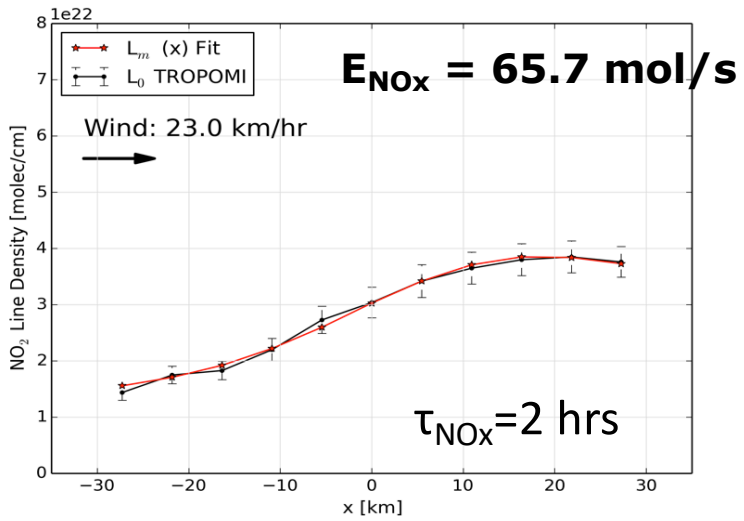




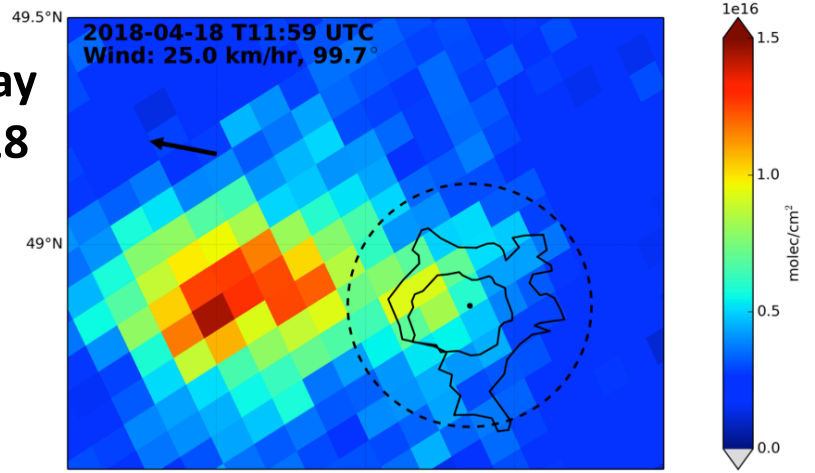


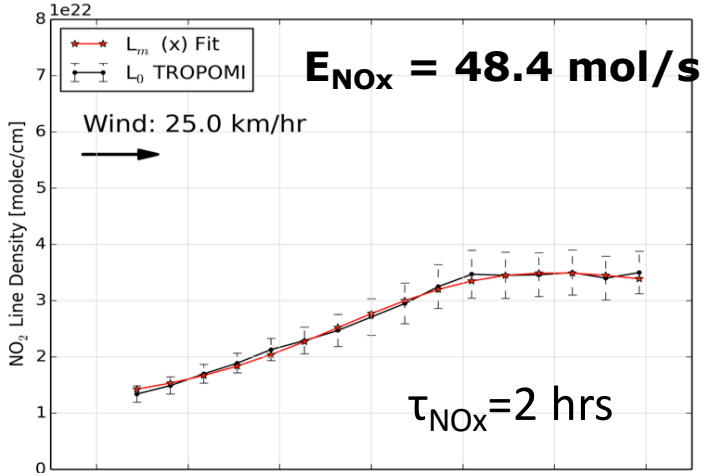
Tuesday 17-04-2018
23 km/h



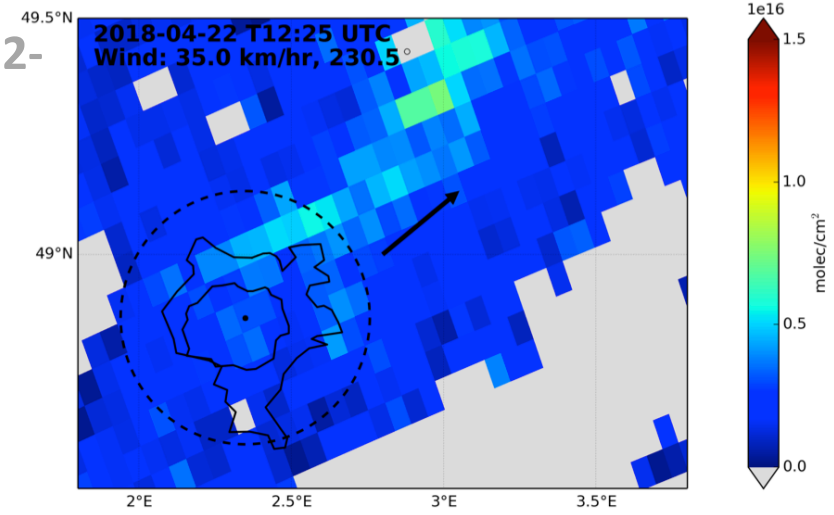


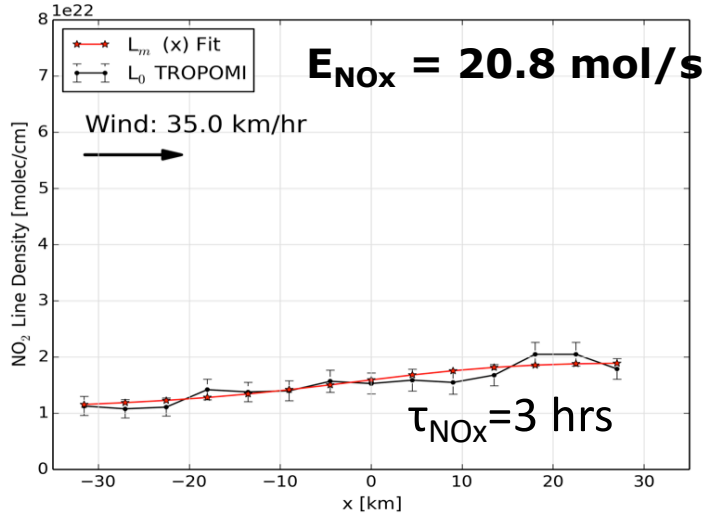
Wednesday 18-04-2018
25 km/h



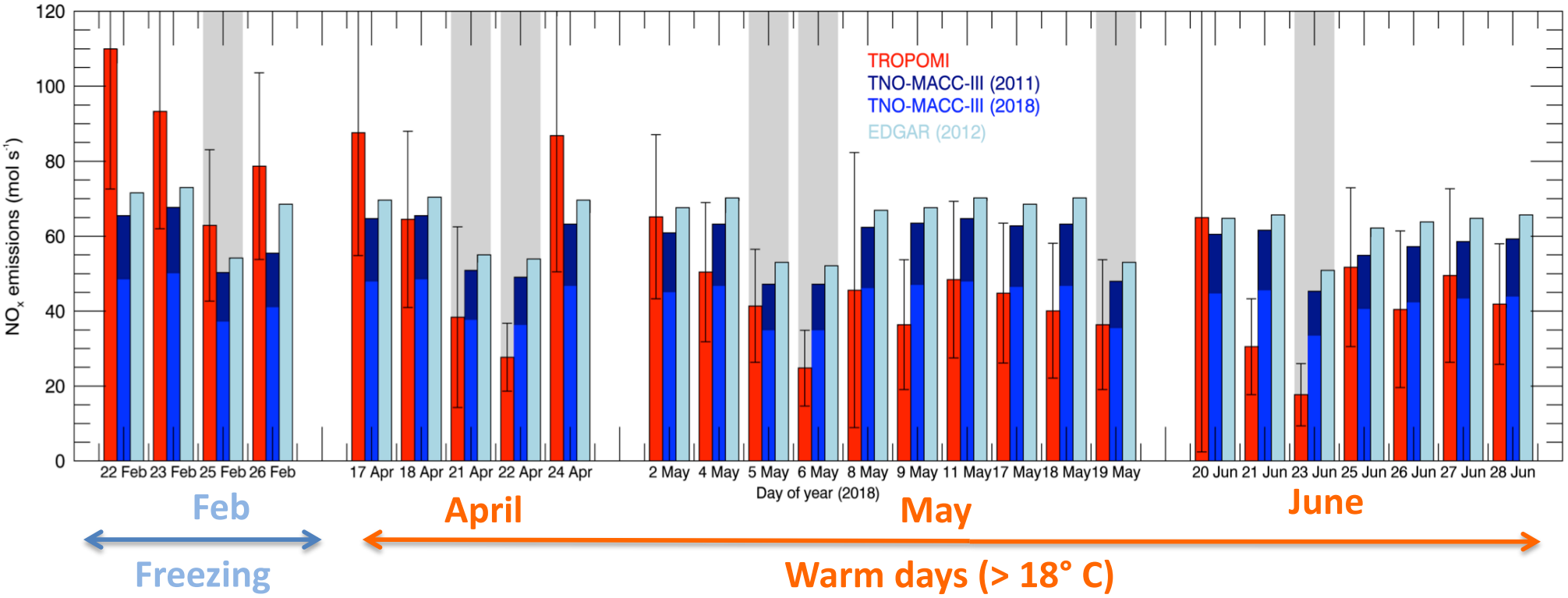


Sunday 22-04-2018
35 km/h





Paris NO_x emissions Feb - Jun 2018

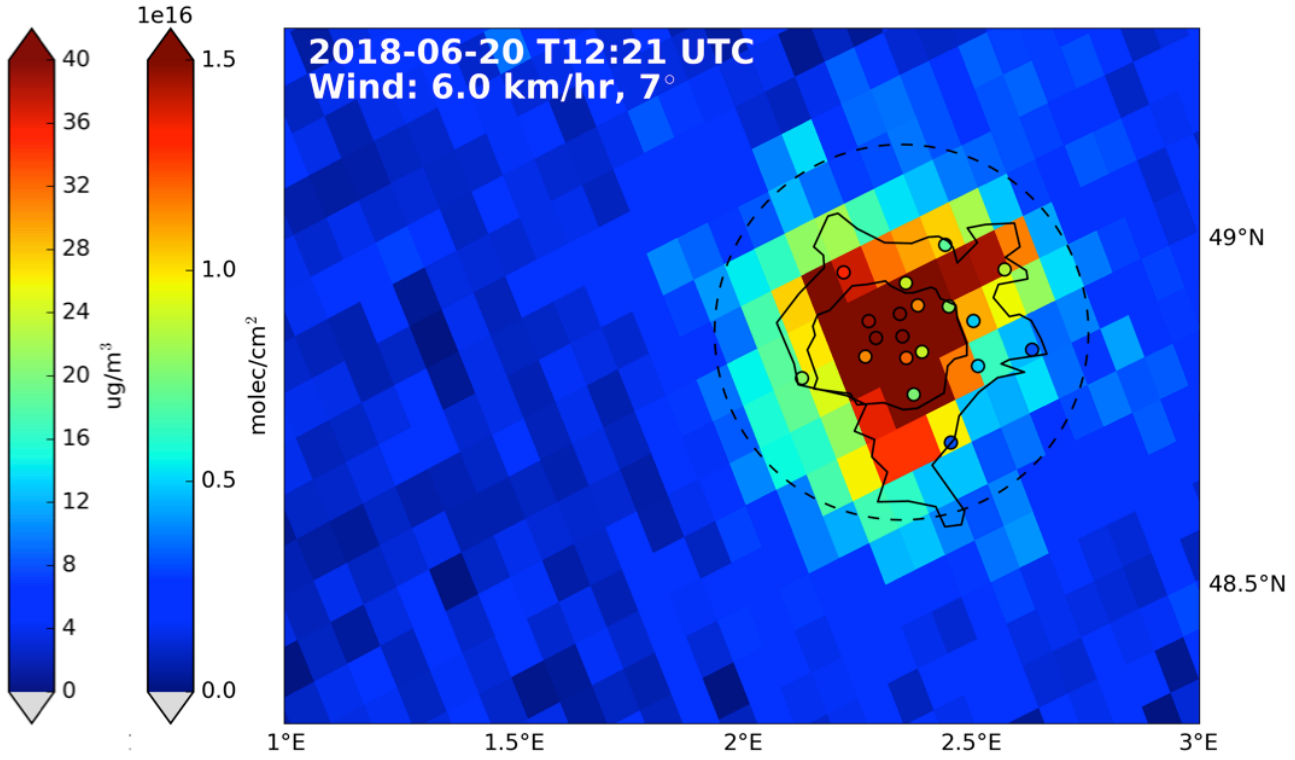


- Daily emission estimates
- TROPOMI captures weekend reductions
- February: TROPOMI higher than inventory
- Apr-June: TROPOMI comparable to TNO-MACC-III (2018)

Summary

- **TROPOMI captures build-up of NO_2 over a source region**
  - Information on spatial distribution of emissions!
- **Estimate noontime NO_x emissions on a day-by-day basis**
  - Weekend effect clearly seen!
- **TROPOMI NO_x emissions 5-15% lower than inventory for 2012**
  - long way from projected 26% reductions for 2018 vs. 2011

Day with stagnation



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Some needs for OMI and TROPOMI NO_2

Lessons learned from the FP7 QA4ECV project

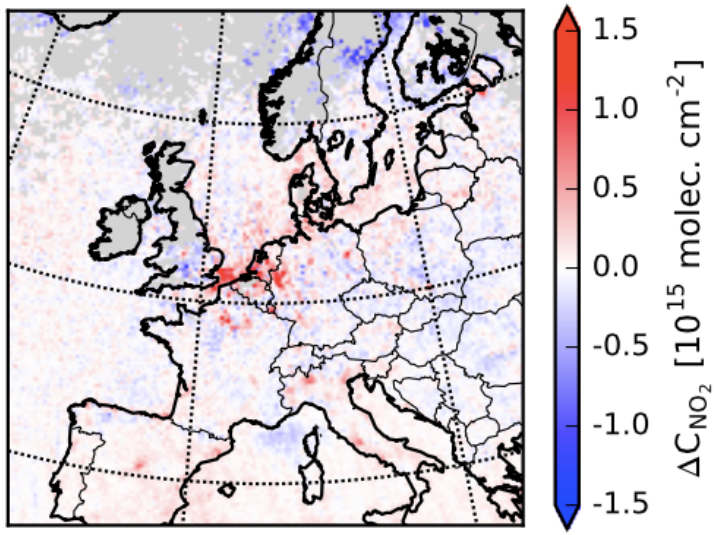
Need pixel-scale information on state-of-the-atmosphere!

- Take surface reflectance anisotropy into account
- This means: replace LER in cloud and NO_2 AMF calculation

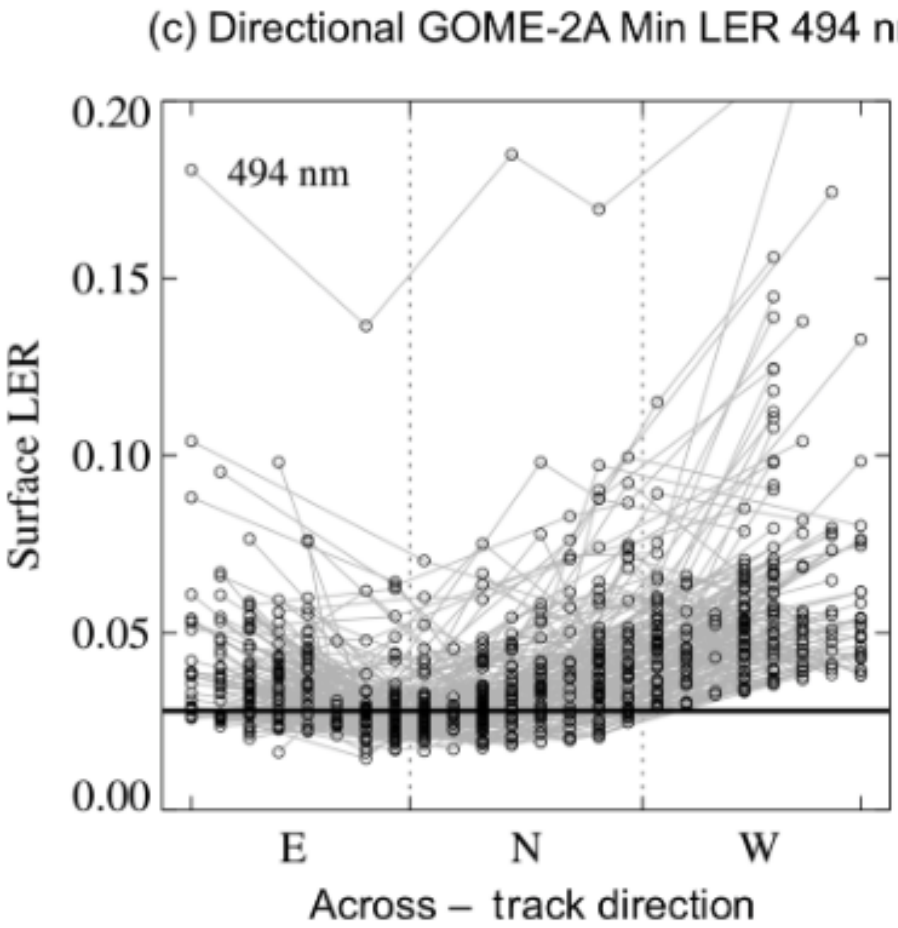
- Pixel-resolution a priori profiles of NO_2
- This means: replace profiles from coarse-scale models

TM5 at 100x100 km^2 → WRF-Chem 20x20 km^2

Visser et al., ACP, 2019



Some needs for OMI and TROPOMI NO_2

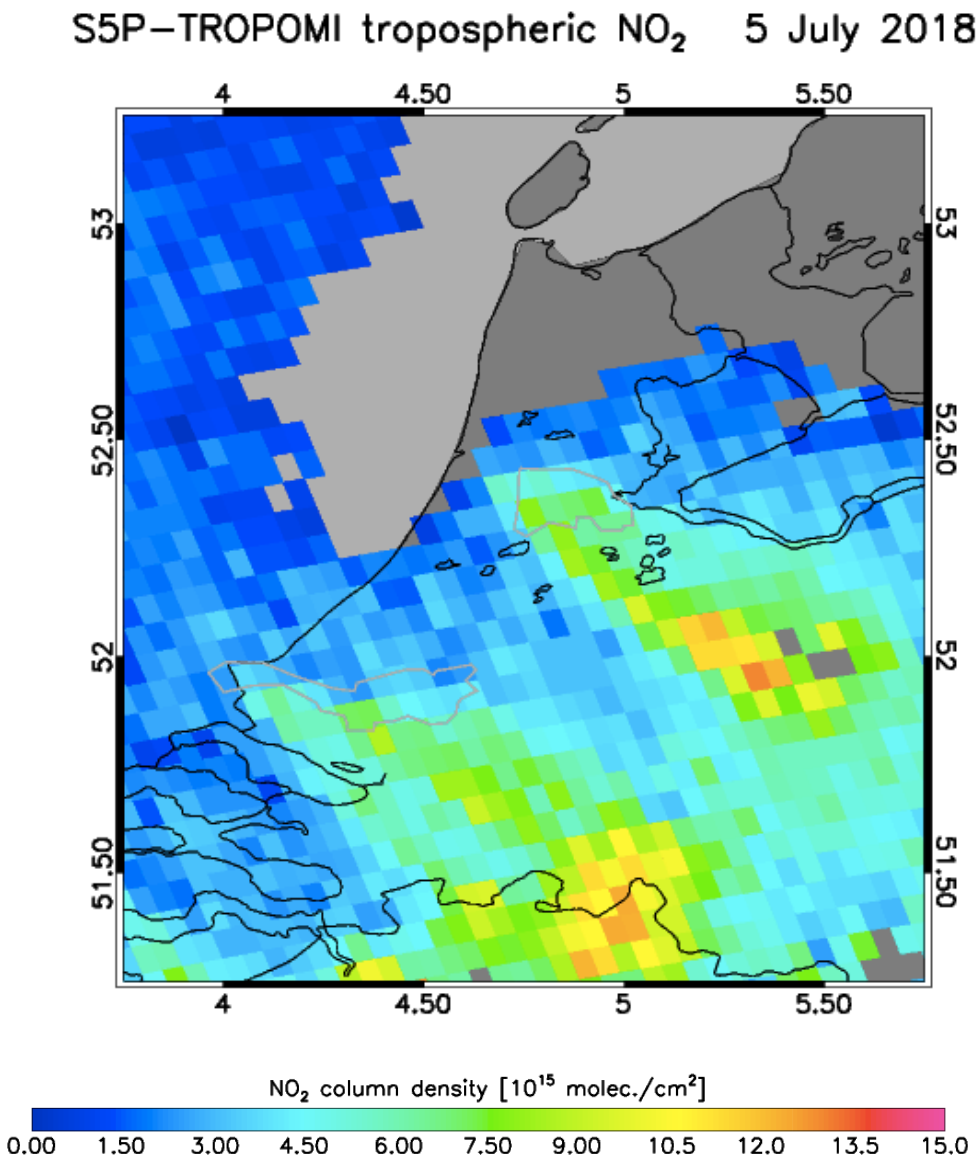


Lorente et al., AMT, 2018

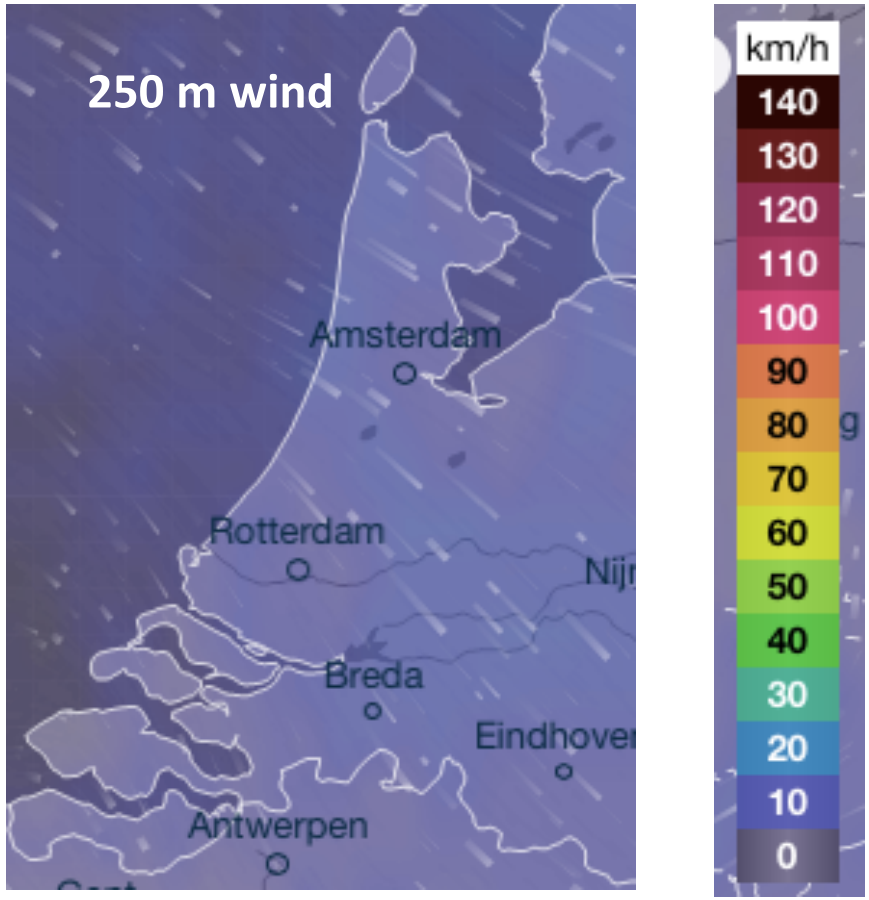
3-D effects of clouds

Foto Tim Vlemmix

Focus on The Netherlands

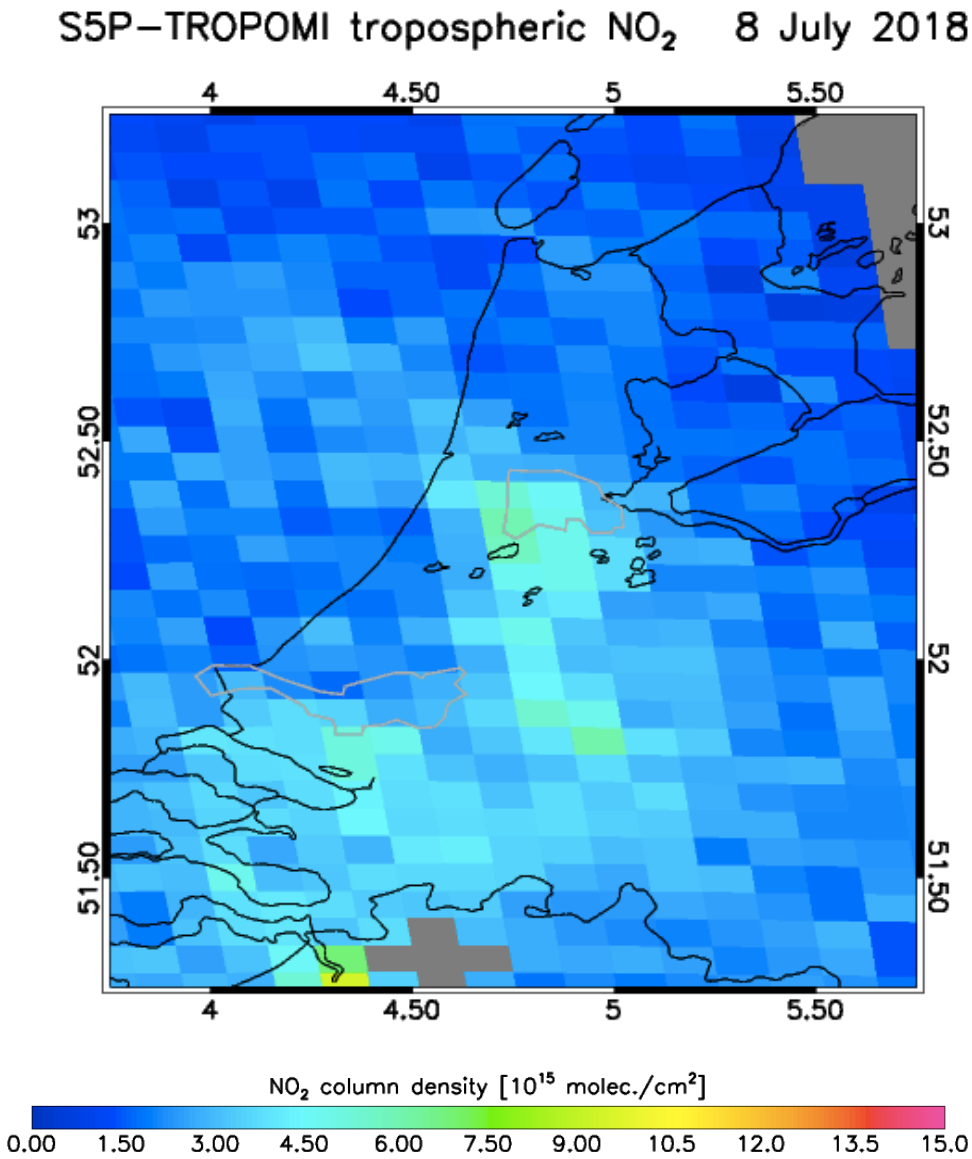


THURSDAY

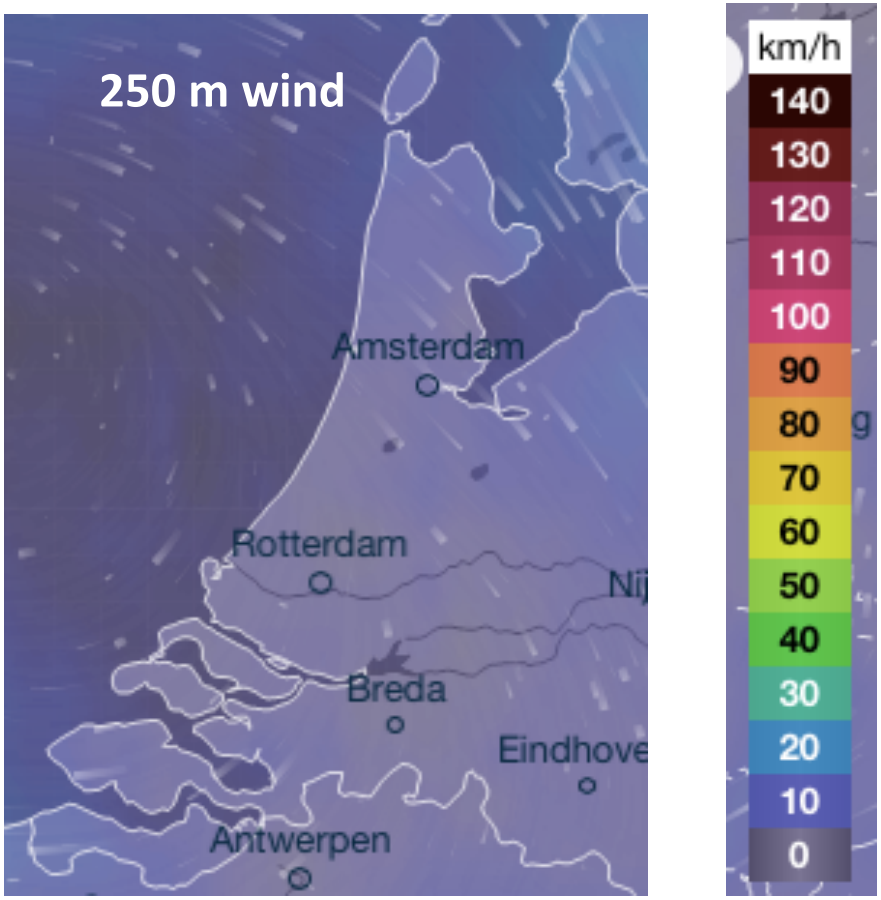


Amsterdam & Rotterdam: 10 km/u

Focus on The Netherlands



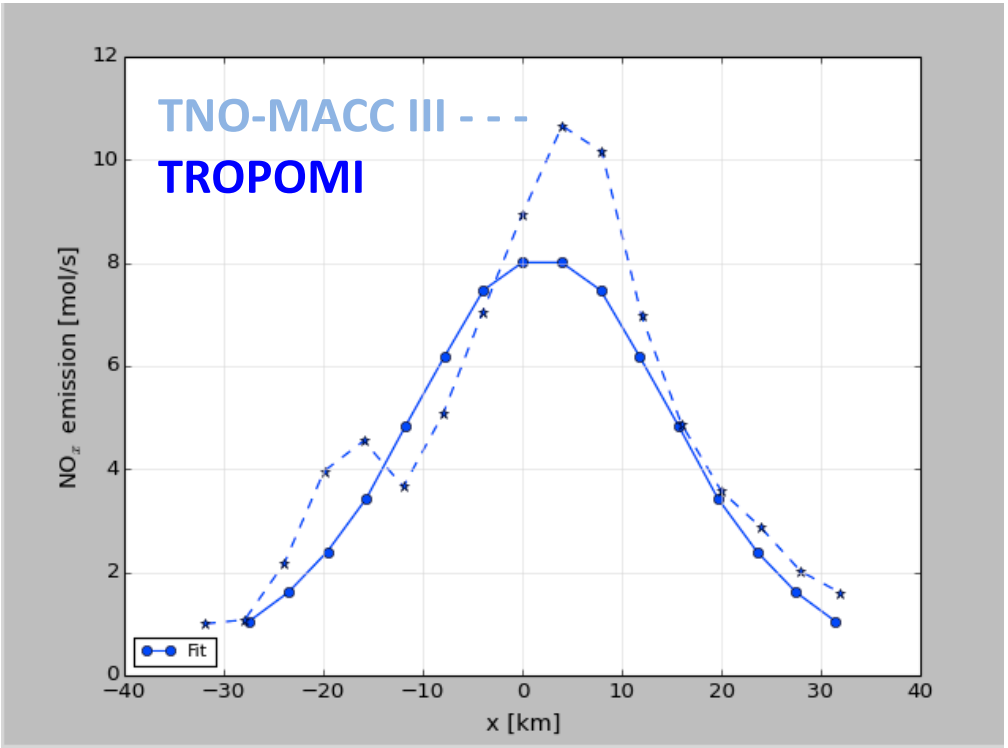
SUNDAY



Amsterdam & Rotterdam: 5 km/u

NO_x emission pattern

Constraint on emission patterns:

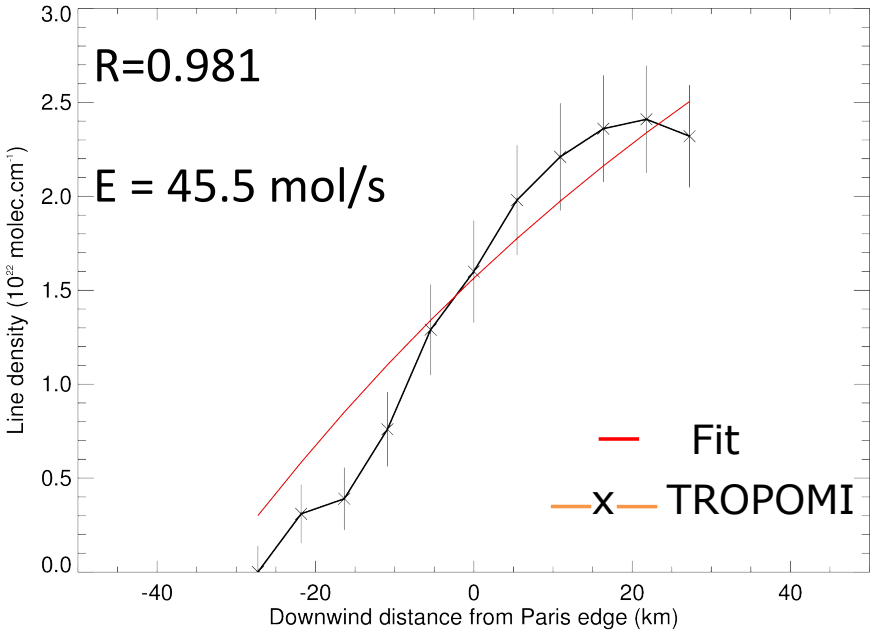


- TROPOMI provides information on the sub-urban distribution of emissions in Paris.
- The spatial variability in our inferred NO_x emissions is similar to the a priori distribution from TNO-MACC-III.

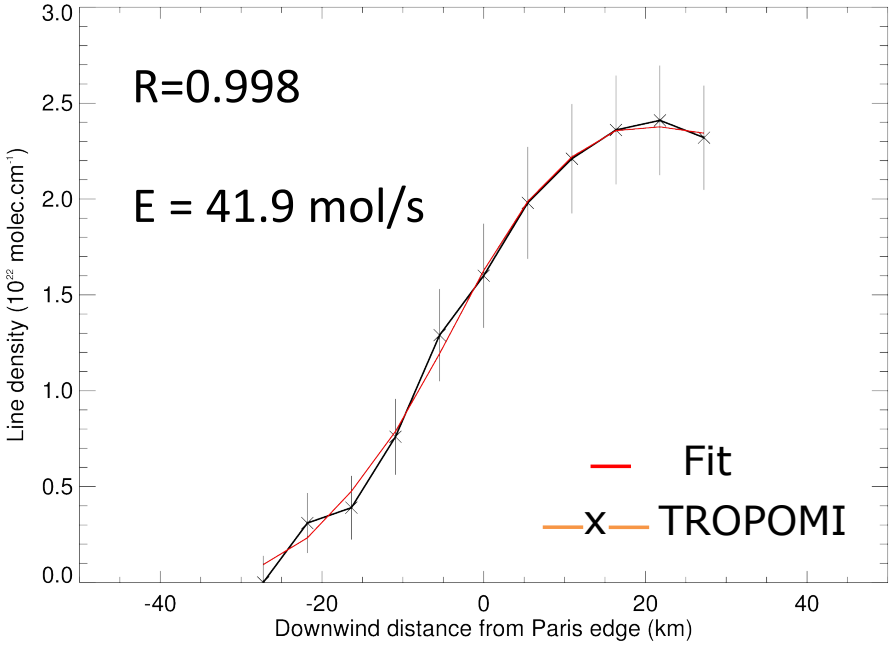
Ensemble of forward model simulations

17th April 2018 T12:18 UTC
Wind 24 km/h , wind_dir = 197°

Levenberg-Marquardt



Best ensemble member



Uncertainties in NO_x emissions

- Replace NO_x:NO2 ratio CAMS by Eiffel Tower: <3% difference
- Wind speed uncertainty of ±20% has similar effect
- Replacing CAMS by CLASS a priori [OH] has some effect
- Weak sensitivity to emission pattern (next slide)

	Uncertainty	Effect on NO_x emissions
S5P-TROPOMI NO_2 column	30%	30%
NO_2:NO_x ratio	20%	<3%
Wind speed	20%	20%
A priori NO_x loss rate	50%	15%
A priori emission pattern	20%	10%
Total uncertainties assuming uncorrelated error contributions		±50%

Discussion

Clear-sky days only, emissions for noon-time
Method requires advection in well-defined direction
(no re-circulation)

OH in Paris BL from CAMS vs. CLASS

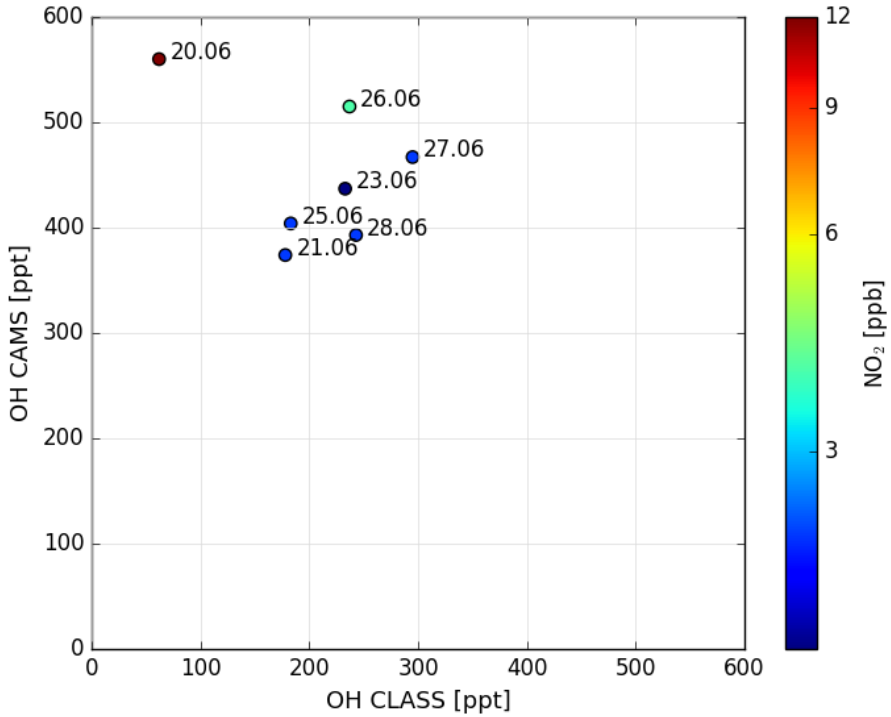
Weak constraint on NO_x lifetimes:

±11 hrs in February

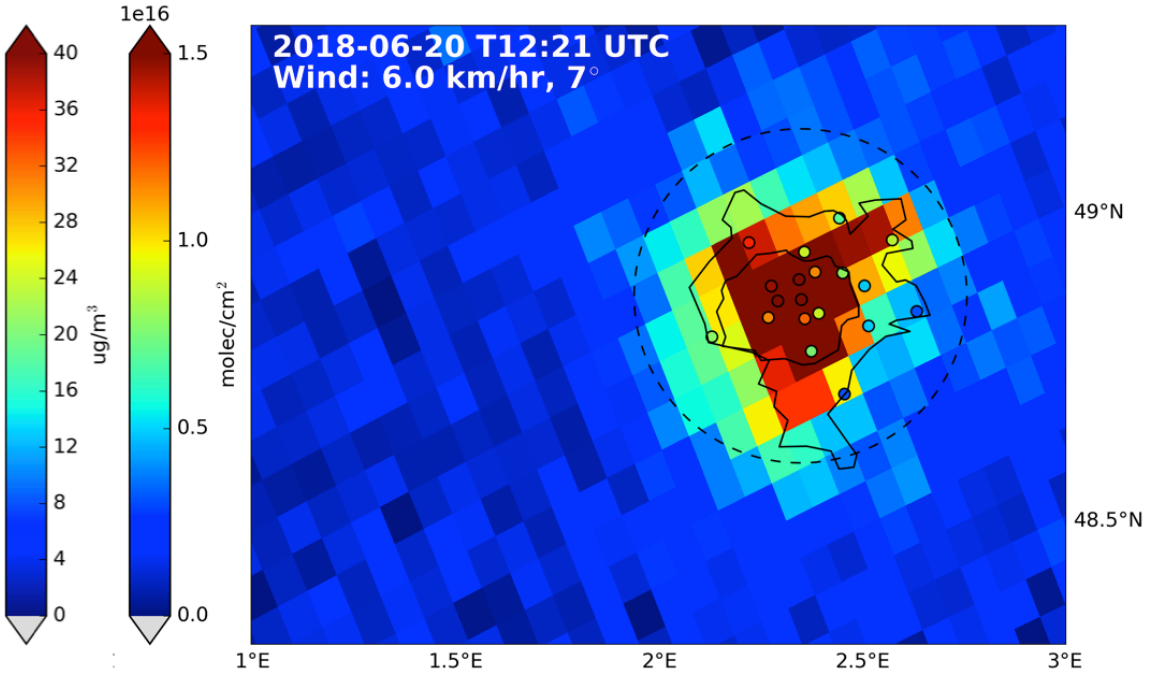
2-4 hrs in April-June

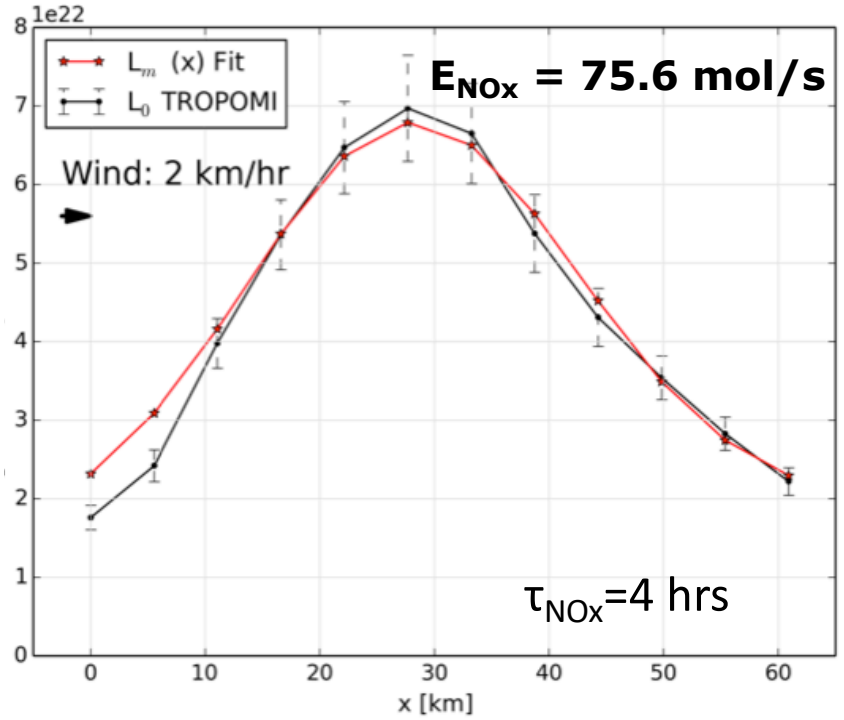
NO_x lifetimes merely represent an improvement to prior, uncertain knowledge on OH concentrations from the CAMS model, constrained via the observed line densities.

The lifetimes correspond to [OH] of 1-12×10^6 molec./cm^3, consistent with other estimates[7,16,24,25].



Day with stagnation





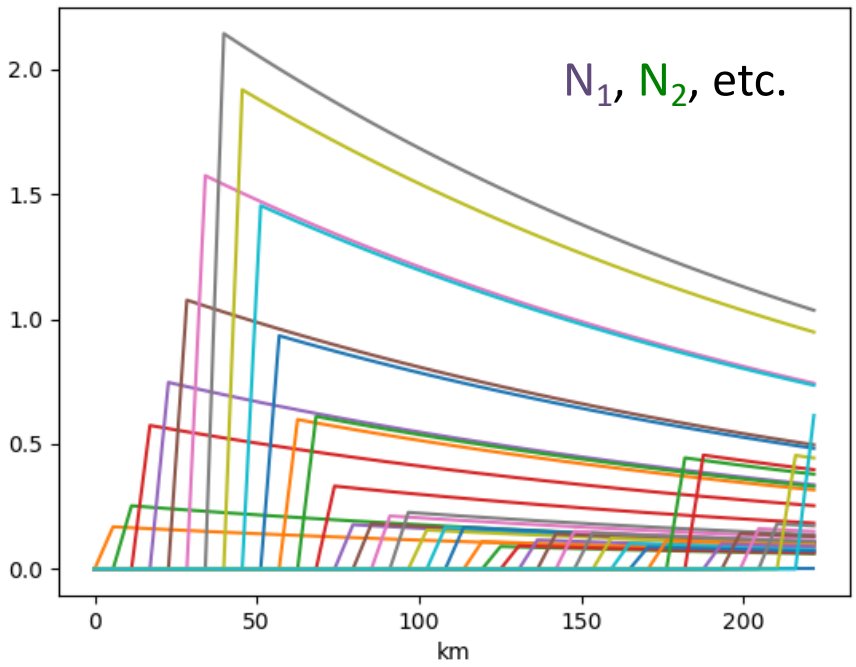
Superposition model (1)

- Forward model function that simulates NO_2 line density as a function of distance *x* over the city.
- Individual *i* representations of the column model:

$$N_i(x) = \frac{E(x_i)}{k}\left(1 - e^{-k(x-x_i)/u}\right)\frac{[\mathrm{NO_2}]}{[\mathrm{NO_x}]}$$

$$k = \frac{k'[\mathrm{OH}][\mathrm{M}]}{([\mathrm{NO_x}]/[\mathrm{NO_2}])}$$

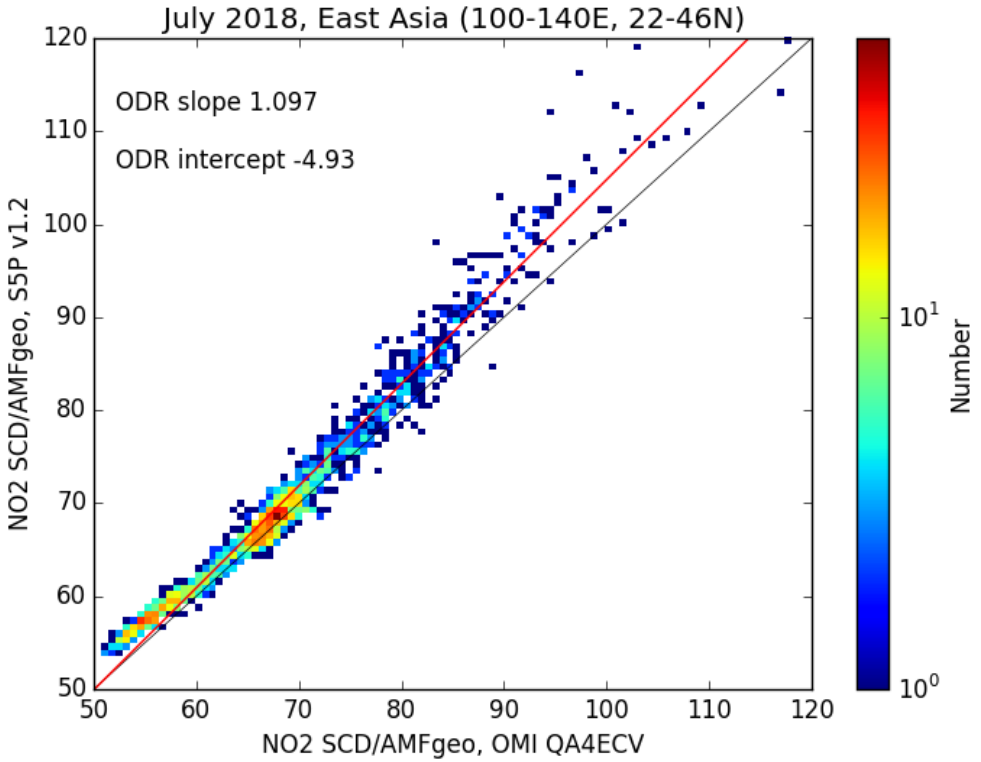
$N_i(x)$
[molec/cm]



Accounts for spatially varying emission rates E_i in the urban area

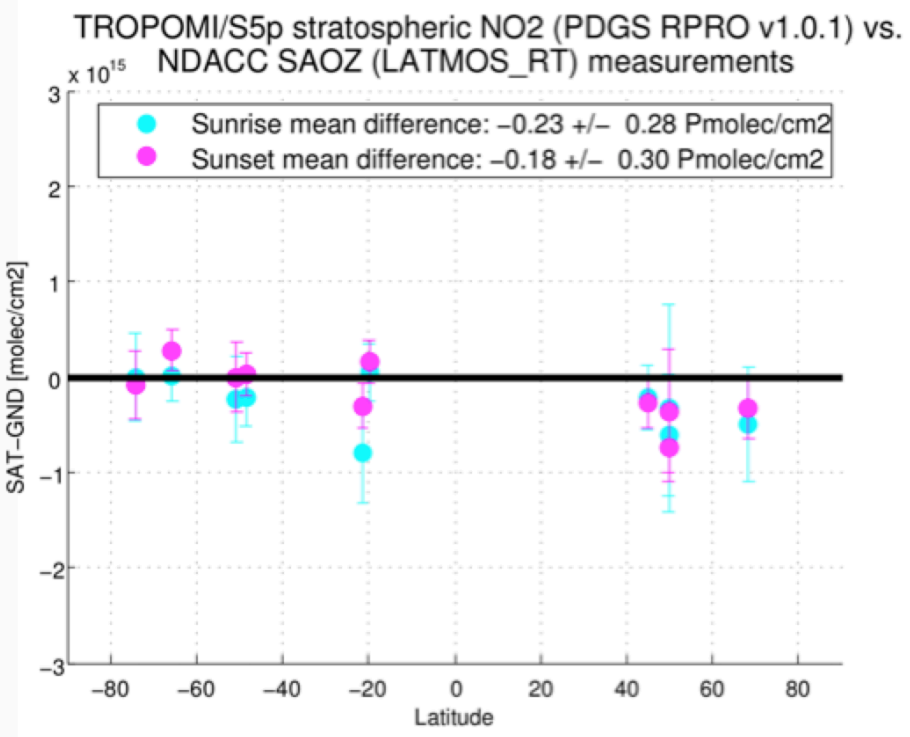
Why is TROPOMI NO_2 biased low?

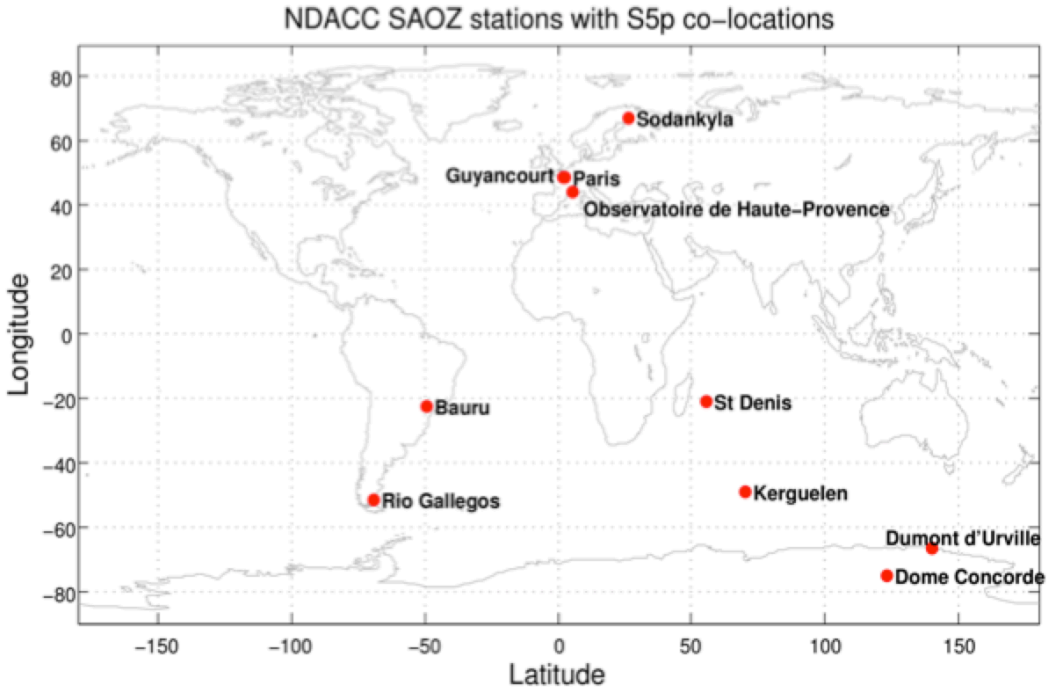
SCD/AMF_{geo}



- TROPOMI SCDs most of the time higher than OMI QA4ECV SCDs

Why is TROPOMI NO_2 biased low?

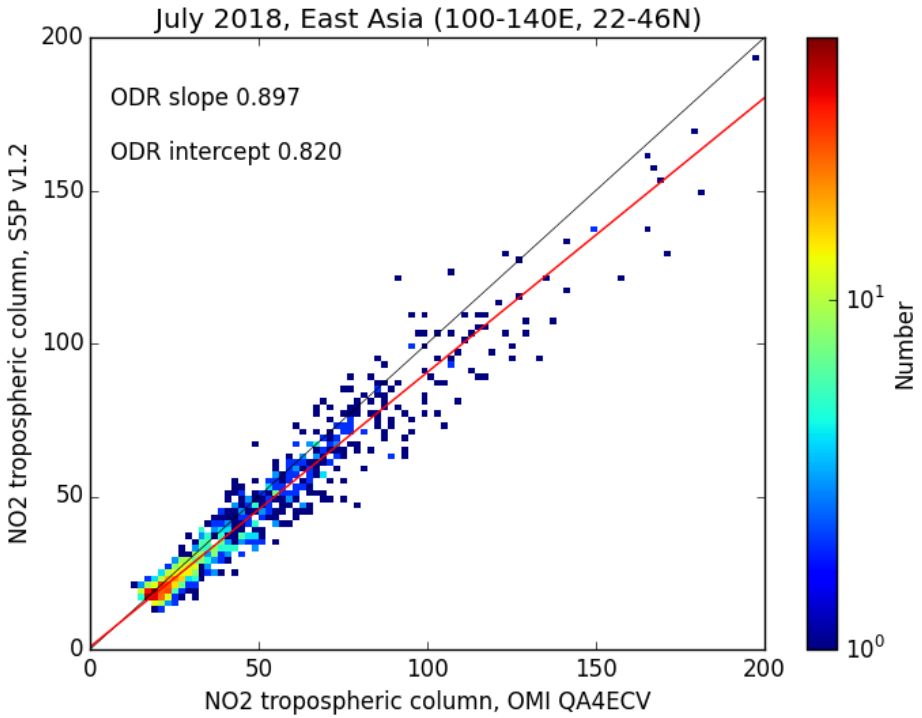




- TROPOMI stratospheric NO_2 similar or lower than SAOZ

Why is TROPOMI NO_2 biased low?

Tropospheric NO_2

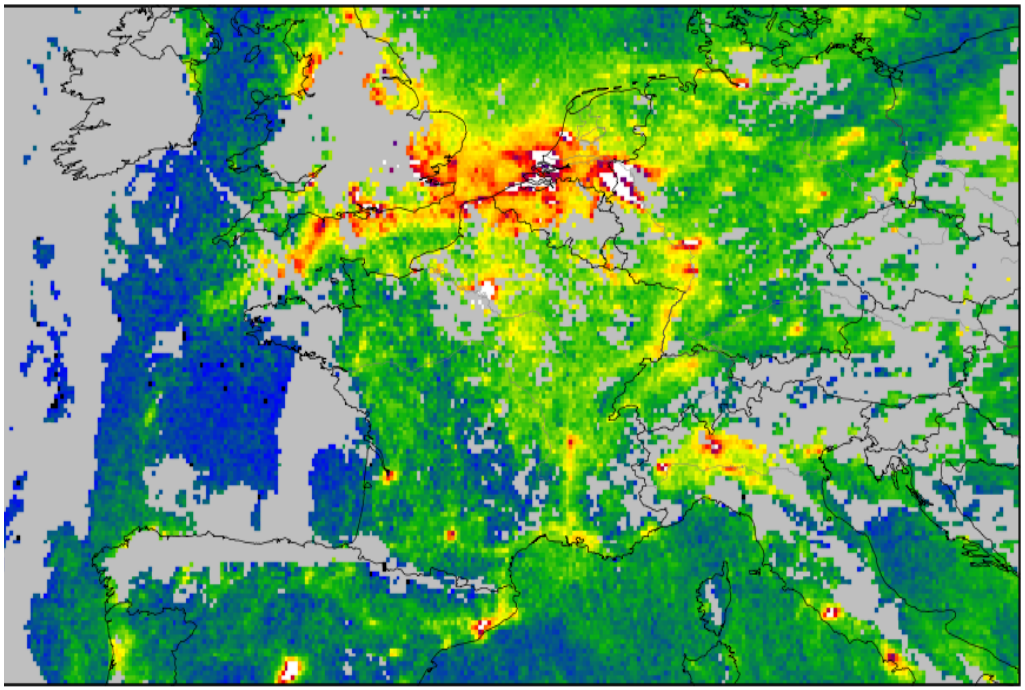


- Tropospheric column lower than OMI QA4ECV
- Validation: 10-50% lower than MAX-DOAS

  Points to errors in the AMF
  - A priori NO_2 profile shape
  - Surface albedo
  - Clouds

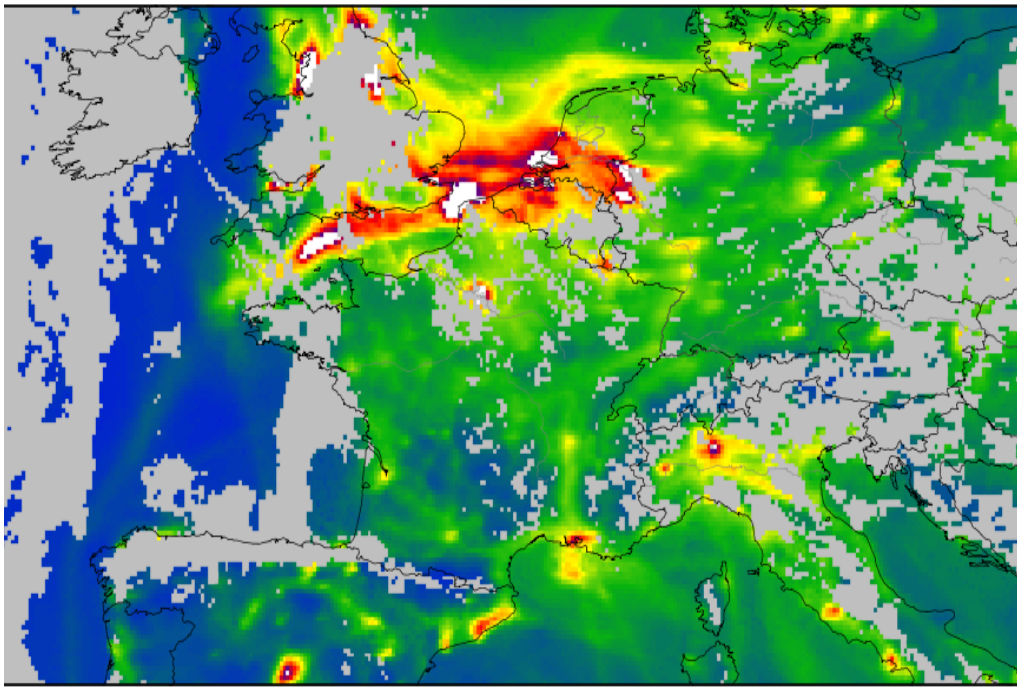
Using a-priori profiles from CAMS-regional

TROPOMI tropospheric vertical column of nitrogen dioxide using CAMS a-priori profile

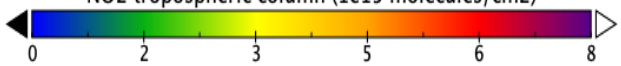


TROPOMI tropospheric vertical column of nitrogen dioxide using CAMS a-priori profile (10^15 molecules/c...

CAMS-rg NO2 tropospheric column, 26 July 2018

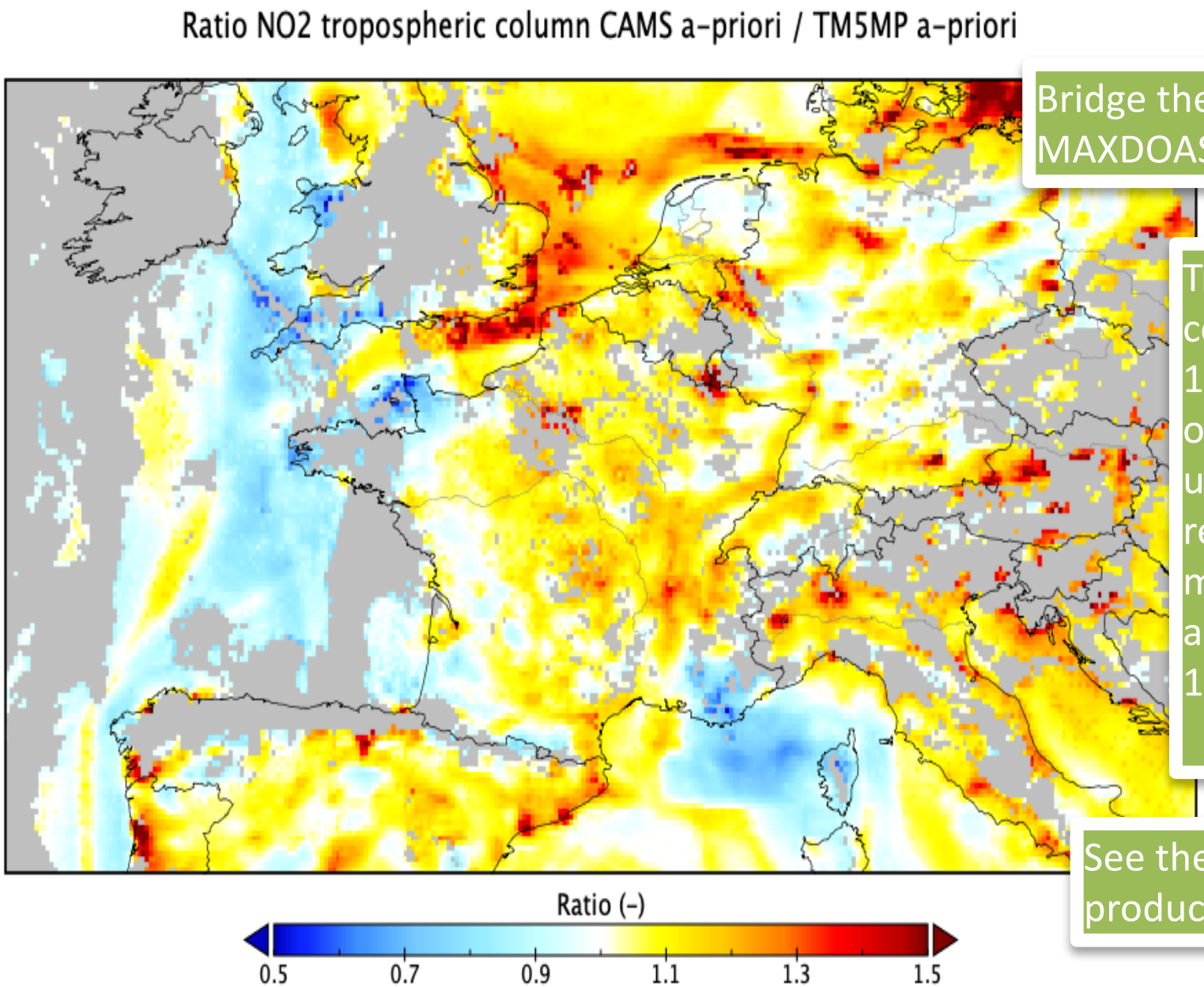


NO2 tropospheric column (1e15 molecules/cm2)



Single overpass, 26 July 2018

Using a-priori profiles from CAMS-regional



Bridge the gap with MAXDOAS

Tropospheric column increases by 10-50% over hotspots when using high-resolution regional model a-priori profiles 1x1 degree -> 0.1x0.1 degree

See the S5P NO2 product user manual